A DIFFUSION MODEL OF AN ISOTHERMAL TUBULAR FLOW REACTOR

by

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INTRODUCTION

The mechanism by which heat or mass is dispersed in tubular flow reactors, and other chemical process equipment through which a fluid is flowing, has recently gained a great deal of attention. In order to solve the problem of determining the conversion and mixing characteristics of a tubular flow reactor it is necessary to consider a rather complicated system of partial differential equations. The system is complicated because the velocity profile is not flat and because there are not only gradients of concentration but also of temperature and pressure. To obtain the general solution of this problem analytically is, of course, out of the question and the numerical solution is tedious and requires a very large computer. At the same time, however, there is the larger question of whether from a practical point of view the effects mentioned above are important enough to justify the effort needed to solve the problem. Therefore, it would be interesting to obtain some estimate of the magnitude of some of these effects.

In an attempt to exploit a technique which might give a partial insight into the behavior of tubular flow reactors, a flat velocity profile and a concentration gradient due to dispersion only is assumed. Earlier investigators introduced the concept of longitudinal diffusion to describe the phenomenon of dispersion in the direction of flow of the mean fluid stream, and the concept of radial diffusion to describe dispersion in the direction perpendicular to the mean fluid flow. This work will be limited to the study of axial dispersion.

A number of models have been proposed for the dispersion phenomenon associated with flow through tubular flow reactors, fluidized beds and packed beds, for example, the researches of Danckwerts (2) and Levenspiel and Smith (12) for flow through tubular flow reactors by the diffusion-type model, the work of Carberry and Bretton (1) for flow through fixed beds by the cell-model, the work of Miyauchi (14) by a cell with back-mixing, and the diffusion-dead space model by Koump (9) and Turner (20). All of these models idealize the three-dimensional problem into a one-dimensional transport equation.

The most commonly used model is the diffusion-type model. This model assumes that the overall rate of axial dispersion is proportional to the bulk concentration gradient of the diffusing component in the axial direction and is measured by the spreading of a concentration wave in an axial direction. This model is also used in this research to investigate the effect of axial mixing on the performance of isothermal tubular flow reactors. With this model, a boundary value problem of a second-order partial differential equation is obtained when a material balance is made with respect to a traceable component in the fluid.

In general, a change in boundary conditions may have a significant effect on the behavior of the solution of a partial differential equation. Hence, in order to avoid the possibility of error due to the improper choice of boundary conditions, one must prove that the boundary conditions used in solving a particular initial or boundary value problem are realistic, or that the solution of the particular differential equation is not sensitive to the choice of boundary conditions.

Various sets of boundary conditions have been suggested for use with the diffusion-type model to predict dispersion phenomenon associated with flow through tubular flow reactor. Danckwerts (2), for example, introduced a set of boundary conditions which satisfy the continuity requirement at both ends of a reactor. Many other investigators introduced sets of boundary conditions which do not conserve the continuity requirement at the inlet of reactor. Hulburt (8), for example, introduced a set of boundary conditions which neglect the diffusion at the inlet of the reactor. Levenspiel and Smith (12) further approximated a reactor of finite length by a reactor of infinite length. Although both of these two sets of boundary conditions are convenient for estimating the relative importance of reaction and diffusion rates, there has been no attempt made to justify numerically the quantitative relationships among the solutions obtained by use of these sets of boundary conditions. Another possible set of boundary conditions which meet the continuity requirement at both ends of the reactor, but hitherto unknown, is investigated in the course of this research.

The present study represents an effort to increase the theoretical knowledge of isothermal tubular flow reactors by investigating the effects of axial dispersion on the performance of a reactor based on a diffusion-type model. The differences and the relationships among the steady-state response curves against the various types of concentration pertubation obtained by use of the different sets of boundary conditions are numerically evaluated. Theoretical response curves of exit concentrations against impulse, step input and frequency response functions are presented in order to illus-

trate the method of determining mean dispersion coefficients from available dynamic experimental data.

LITERATURE SURVEY

As indicated in the previous section, different models of fluid flow and various sets of boundary conditions have in recent years been proposed in order to predict the effects of axial mixing on the performance of such process equipment as packed beds, fluidized beds and tubular flow reactors. Table 1 summarizes the work of previous investigators. Included in the table are the mathematical models and boundary conditions employed, the process equipment and experimental technique used, and the method of analysis of the data.

Danckwerts (2) presented a general discussion of how distribution functions for residence time can be defined and measured for pipes, packed beds and reactors from dynamic step-input experiments. He also reported the performance of a steady-state flow reactor based on the diffusion-type model with a first-order chemical reaction using a set of continuity preserving boundary conditions at both ends of the reactor. The effect of axial mixing on the first-order conversion is also compared with that of a plug flow reactor. The boundary conditions Danckwerts introduced will be denoted as B.C.(I).

Hulburt (8) investigated steady-state first-order kinetics using a diffusion model to study the effect of axial dispersion on the conversion. He presented a set of boundary conditions which neglect the effect of diffusion at the reactor entrance. This set of boundary conditions will be denoted as B.C.(II).

An analysis of these two boundary conditions was presented by Wehner and Wilhelm (24) for a steady-state flow reactor with axial diffusion and first-order reaction. They have indicated that the Hulburt solution (B.C.II) is convenient for estimating the relative

Table 1. Summary of previous investigations.

Investigators	Models	B.C.	Equipment	Experimental Technique	Method of Analysis
Danckwerts (2)	Diffusion-type	(I)	Reactors Blenders Packed tubes	Step input	
Kramers, et. al. (10)(11)	Diffusion-type	(I)(III)	Cylinder	Frequency response	
Levenspiel and Smith (12)	Diffusion-type	(III)	Reactors	No experiment	Variance, σ^2
Head, Hougen, and Walsh (7)	Diffusion-type	(III)	Tube	Impulse	Fourier Transforms
Ebach (4)	Diffusion-type	(III)	Packed beds	Frequency response; Pulse functions	Point source
Otake and Kunugita (16)	Diffusion-type	(II)	Packed beds	Step input	Approximate solution
Hulburt (8)	Diffusion-type	(II)	Reactors	Steady-state	
Deisler and Wilhelm (3)	Diffusion-type	(III)	Catalytic Reactors	Frequency response	Amplitude alone
Yagi and Miyauchi (25)	Diffusion-type	(I)	Reactor Fluidized bed	Impulse; Frequency response	Point source

Table 1. (con't.)

Investigators	Models	B.C.	Equipment	Experimental Technique	Method of Analysis
Strang and Geankoplis (18)	Diffusion-type	(II)	Packed bed	Frequency response	Amplitude alone
McHenry and Wilhelm (13)	Diffusion-type	(III)	Packed beds	Frequency response	Amplitude alone
Vincent (22)	Diffusion-type	(III)	Heat exchanger	Frequency response	
Romano and Pigford (17)	Diffusion-type	(III)	Fluidized beds	Frequency response	Fourier Transforms
Carberry and Bretton (1)	Cell-model	(III)	Fixed beds	Impulse	
Miyauchi (14)	Cell with back-mixing	General discussions		Step Impulse	
Turner (20)	Diffusion-dead space model	None	Packed beds	Frequency response	
Koump (9)	Diffusion-dead space model	(III)	Packed beds	Step input Frequency response Impulse	
von Rosenberg (23)	Diffusion-dead space model	(III)	Packed beds	Step input	

importance of reaction and dispersion rates, but is not consistent with the limit of a stirred tank reactor with infinite diffusivity in the reaction section. They have investigated the validity of these boundary conditions by simultaneous solution of three differential equations for the reaction section, fore and after sections. This investigation was based on the fact that axial dispersion may occur in the latter two sections.

Levenspiel and Smith (12) provided an anlytical soltuion to the problem of fluid mixing in a continuous flow system without chemical reaction using the diffusion-type model but with a different set of boundary conditions. They presented a set of approximate boundary conditions by assuming that the effect of diffusion at the reactor entrance is negligible so that a discontinuity exists at this boundary. They further approximated a reactor with finite length by a reactor with infinite length. Based on this set of boundary conditions they showed two methods, a single point method (maximum concentration) and a method based on the variance of the concentration distribution (σ^2), for evaluating the axial dispersion coefficient from experimental measurements. They also worked out some examples to determine the conditons of applicability of the model. This set of boundary conditions will be denoted as B.C. (III).

In a short note, van der Laan (21) discussed an aspect of the work reported by Levenspiel and Smith (12). While Levenspiel and Smith (12) treated the diffusion-type approach by restricting the solutions to infinite lengths, van der Laan (21) treated the diffusional type flow in a more general manner so as tc include the

case of finite length and of varying dispersion by applying suitable boundary conditions. He treated the general solution by means of a Laplace transformation and avoided the back transformation by applying momentum analysis, that is, the method of variance of the concentration distribution. An illustration was given for the evaluation of the dispersion coefficient using the results of calculations of mean residence time and variance.

One of the pioneer investigations of axial dispersion in process equipment was that of Kramers and Alberda (10). They investigated axial dispersion for the flow of water through a bed of Raschig rings. A frequency response technique was used, and the results were analyzed on the basis of the assumption that the rate of axial dispersion can be described by a diffusion-type model with B.C.(I). In order to avoid the complexities involved in obtaining an exact analytical solution, they have suggested that the performance function of the system be approximated by n completely mixed vessels connected in series. The feasibility of this approximation is open to question, except for the system involving a small degree of dispersion. Kramers, Honig, and Croockweit (11) extended their work to the study of longitudinal dispersion in liquid flow by using B.C.(II). Actual derivation of the analytical solution, however, was carried out by means of B.C.(III).

Yagi and Miyauchi (25) investigated the operational characteristics of continuous flow reactors in which the reactants are mixing and described the dispersion phenomenon in terms of mean longitudinal dispersion applying the diffusion-type model and B.C.(I). An impulse function was used, and the results were analyzed by means

of a single point method (i.e., maximum points). The data of Ebach (4) on impulse experiments were analyzed on the same basis as those of Yagi and Miyauchi (24). It is known that the use of a single datum in drawing conclusions for an entire experiment may lead to serious error because of the probable experimental error. Although the data shown in the work of Ebach (4) gave close agreement between impulse experimental values and calculated values at low degree of dispersion, the feasibility of using peak values of their impulse data at high degree of dispersion leaves much to be desired.

In experiments with the frequency response technique, Ebach (4) used amplitude data alone to analyze the mean dispersion coefficient. He stated that the axial dispersion coefficient calculated from phase shift data gave poor accuracy and reproducibility. The reasons for this fundamental fault were clearly illustrated in his paper. Strang and Geankoplis (18) studied the axial dispersion of liquids in packed beds by the same frequency response technique. The diffusion model was used, and the data were also analyzed using amplitude data alone.

Deisler and Wilhelm (3) also employed the frequency response technique to investigate both pore diffusion within particles and longitudinal dispersion in the fluid between particles in the laminar flow of gas mixtures. A diffusion-type model was used, and the analytical solution was derived based on B.C.(III). The axial dispersion was evaluated simultaneously with the diffusivity in solid and the adsorption constant. Applying a similar model, boundary conditions and experimental technique, McHenry and Wilhelm (13) studied the axial dispersion of binary gas flowing through beds of

inert spherical particles. The results were evaluated using amplitude data alone.

A very interesting paper was published by Head, Hougen and Walsh (7) in their study of the axial dispersion of water flowing through a vertical tube. A diffusion-type model was assumed together with B.C.(III). Experimental data were obtained by the pulse wave technique, and the results were analyzed by reducing pulse data to the frequency response form by means of a Fourier transformation. The reduced data were plotted in a Bode diagram to evaluate the mean dispersion coefficient by fitting experimental data and calculated values. A similar method of analyzing experimental data was reported by Romano and Pigford (17) in their study of the performance of a fluidized bed by the frequency response technique. The data obtained were reduced to pulse form by a Fourier transformation, and the resulting residence time distribution curve was used to study the mechanism of gas mixing in a fluidized bed, and the effects of gas velocity and bed height on the distribution function.

Otake and Kunugita (15) presented a general discussion of mixing characteristics of reactors without chemical reaction based on a diffusion-type model. Analytical solutions corresponding to the different sets of boundary conditions were presented in their paper for various types of transient inputs. In another paper (16), they investigated the performance of irrigated packed towers using a diffusion-type model and B.C.(II). A step signal was used, and the data were analyzed under the assumption that when the degree of mixing is very small their analytical solution, which is in the form of an n-th term expansion, can be simplified to only the first term.

In the previous section, it was pointed out that many other mathematical models have been proposed to predict the behavior of continuous flow systems. One of the most widely used models beside the diffusion-type model is the diffusion-dead space model. This model for axial dispersion postulates the existence of a stationary fluid region in the voids of the process equipment which can exchange mass with the moving fluid. In a purely theoretical investigation, Turner (20) presented a model of a packed bed which can be classified under the diffusion-dead space category. He introduced a concept hydrodynamically equivalent to the packed bed, which consists of a number of identical channels through which the fluid is flowing. Each of these channels has a number of "pockets" of stationary fluid which are arranged along the main channel. The fluid within the "pockets" is regarded as stationary. The mass transfer between the flowing fluid and the "pockets", and the mass transfer within the "pocket" volume is assumed to take place by molecular diffusion. The model proposed by Turner (20) appears to be too complex to be of practical use. In an attempt to simplify the model, Koump (9) presented the diffusion-dead space model under the assumptions that the axial dispersion of mass takes place by a combination of two mechanisms: a diffusion type process and a mass exchange between the moving fluid and the fluid within the stagnant void volume. The experiment was carried out by three different techniques: frequency response, impulse, and step inputs. The results were analyzed by both the diffusion-type model and the diffusion-dead space model.

Von Rosenberg (23) used the diffusion-dead space model to investigate the problem of the displacement of a fluid from a bed of inert solid by another fluid. He regarded the packed bed as an assembly of capillaries and used the solution of G. I. Taylor (19) for the case of negligible forward and rapid lateral diffusion as a basis of the analysis. He assumed that the axial dispersion takes place by the combined effects of velocity distribution in the pores and rapid diffusion in the radial direction. The concept of an axial dispersion coefficient, therefore, does not enter the picture in this approach.

Other models which are often used in the study of axial mixing are the cell-model and cell with back-mixing. The concept of the cell-model implies that the spread of injected tracer as a result of passage through a bed can be described by the equation of concentration-time behavior in the effluent of the n-th perfect mixing cell. The cell with back-mixing differs from the cell-model in that each one of the n perfect mixing cells is associated with a definite back-mixing.

Carberry and Bretton (1) investigated the longitudinal dispersion during the flow of water through beds of rings by the cell-model. Two different types of input signals were used in their work: an impulse function which injected the tracer instantaneously, and a rectangular input function which injected the tracer within a small given time. In both cases, the results were analyzed by fitting the shape of the experimental response curves to the calculated response curves based on the cell-model. Miyauchi (14) presented a purely theoretical analysis of the cell with back-mixing model.

It seems evident from this literature survey that the diffusion-type model has been employed almost exclusively because, to a first approximation, it gives a reasonable description of the phenomenon associated with mixing inside reactors. There seems to be, however, some disagreement on the method of analyzing experimental data and on which sets of boundary conditions to apply. All of the mathematical solutions are most certainly oversimplified and unrealistic from the standpoint of representing the true mechanism. Due to the simplification involved in the mathematical treatment on B.C.(III), it has been used most often without much quantitative justification.

THEORY

The purpose of this section is to derive the partial differential equation for the diffusion-type model of axial dispersion considered in this investigation, and to present the mathematical expressions for four sets of boundary conditions.

Diffusion Model

This model assumes that the rate of dispersion is proportional to the bulk concentration gradient of the diffusing component in the tubular reactor. Under this assumption, a material balance with respect to a traceable component in the fluid with chemical reaction occurring gives the following conservation of mass equation in vector form:

$$\frac{\partial C}{\partial t} + \text{div}(-D \text{ grad. } C) + \text{div}(\vec{u}C) + r(C) = 0 \qquad (1)$$

where D is a dispersion coefficient or apparent diffusivity which characterizes the dispersion of fluids flowing with a velocity of $\vec{u}$, and C and t represent the concentration and time variables respectively. The first term of equation (1) represents the change in concentration with respect to time. The second term represents the net out-flow of component due to dispersion. The third term represents the velocity gradient while the fourth term represents the rate of chemical reaction.

The following assumptions are made:

1) Only the dispersion in the axial direction, z, is significant, that is, the physically three-dimensional problem of the

second term in equation (1) is idealized to a one-dimensional transport equation.

2) The coefficient D, in equation (1) is independent of position and concentration gradients as in the case of fully developed turbulent flow, and is characterized by $\bar{D}$.

3) Changes in volume are assumed not to take place, so that the velocity, $\bar{u}$, in equation (1) is the same at all cross-sections, and is characterized by $\bar{u}$.

4) Only the chemical reaction of first-order is considered.

With these simplifications, equation (1) becomes the following partial differential equation:

$$\frac{\partial C}{\partial t} = \bar{D} \frac{\partial^2 C}{\partial z^2} - \bar{u} \frac{\partial C}{\partial z} - kC \qquad (2)$$

where k is the reaction rate constant.

Equation (2) is a second-order linear partial differential equation which describes the dispersion phenomenon in terms of time, position and dispersion coefficient. As stated in the previous section, the solution of equation (2) depends on the initial and boundary conditions.

Boundary Conditions

Various boundary conditions have been suggested for use with equation (2). As stated in the previous section, boundary conditions may have a significant effect on the solution of equation (2). Hence, the boundary conditions used with equation (2) must be realistic and quantitatively justified. One of the purposes of this investigation is to find boundary conditions compatible with the solution

of equation (2).

Danckwerts (2) introduced a set of intuitive boundary conditions based on the equation of continuity. He proposed that, owing to the dispersion taking place, the concentration of reactant in the entering stream, C_1, must be larger than the concentration just within the entrance of the reactor.

This was expressed mathematically as:

$$\bar{u}C_1 = \bar{u}C_{z\to 0^+} - \bar{D}\left(\frac{\partial C}{\partial z}\right)_{z\to 0^+} \tag{3}$$

At the outlet of a finite tubular reactor, he proposed the same relationship as equation (3):

$$\bar{u}C_{z\to L^-} = \bar{u}C_{z\to L^+} - \bar{D}\left(\frac{\partial C}{\partial z}\right)_{z\to L^+} \tag{3a}$$

The equation of continuity, however, necessitates equation (3a) to be expressed as:

$$\bar{u}C_{z\to L^-} = \bar{u}C_{z\to L^+} \tag{3b}$$

Substitution of equation (3b) into equation (3a) yields:

$$\bar{D}\left(\frac{\partial C}{\partial z}\right)_{z\to L^+} = 0 \tag{3c}$$

Since $\bar{D}$ is different from zero, the term in parenthesis of equation (3c) has to satisfy the following expression:

$$\left(\frac{\partial C}{\partial z}\right)_{z=L} = 0 \tag{4}$$

Equations (3) and (4) will be designated as B.C.(I).

In an attempt to approximate the above B.C.(I), Hulburt (8) proposed a set of boundary conditions which neglect the effect of diffusion at the reactor entrance. Under this assumption, the second term in the right-hand side of equation (3) vanishes, and the following boundary condition at the inlet of the reactor can be obtained:

$$\bar{u}C_1 = \bar{u}C_{z\to 0^+} \tag{5}$$

At the outlet, z = L, he used the same boundary condition proposed by Danckwerts. Equations (5) and (4) will be designated as B.C.(II).

Levenspiel and Smith (12) and several other investigators (14) (15) proposed a set of approximate boundary conditions. They not only neglected the effect of diffusion directly within the entrance of the reactor, but also treated a finite length of reactor as an infinite reactor, i.e., one value of the dispersion coefficient applied to the entire length and cross-section. This infinite length reactor boundary condition is expressed mathematically as:

$$\lim_{z\to\infty} C(z,t) = 0 \tag{6}$$

Equations (5) and (6) will be designated as B.C.(III).

Another set of boundary conditions which has never been employed previously is presented in this investigation. This set of boundary conditions preserves continuity just as those of Danckwerts. The only approximation made in this set of boundary conditions is that the finite length of reactor is treated as a section of an infinitely long reactor as in the case of B.C.(III). The feasibility

of this set of boundary conditions will be discussed in a later section in connection with steady-state first-order conversion and the response to various transient inputs. Equations (3) and (6) will be designated as B.C.(IV).

Table 2 summarizes the proposed boundary conditions for use with equation (2).

Table 2. Summary of proposed boundary conditions.

Boundary Conditions	Inlet ($z = 0$)	Outlet ($z = L$)
B.C.(I)	$\bar{u}C_1 = \bar{u}C_{z\to 0^+} - \bar{D}\left(\frac{\partial C}{\partial z}\right)_{z\to 0^+}$	$\left(\frac{\partial C}{\partial z}\right)_{z=L} = 0$
B.C.(II)	$\bar{u}C_1 = \bar{u}C_{z\to 0^+}$	$\left(\frac{\partial C}{\partial z}\right)_{z=L} = 0$
B.C.(III)	$\bar{u}C_1 = \bar{u}C_{z\to 0^+}$	$\lim_{z\to\infty}(z,t) = 0$
B.C.(IV)	$\bar{u}C_1 = \bar{u}C_{z\to 0^+} - \bar{D}\left(\frac{\partial C}{\partial z}\right)_{z\to 0^+}$	$\lim_{z\to\infty}(z,t) = 0$

MATHEMATICAL TREATMENT

The solutions of equation (2) which satisfy the four sets of boundary conditions tabulated in Table 2 of the previous section are derived by means of Laplace transformations. This has the advantage of transforming a partial differential equation into an ordinary linear differential equation. To derive the expressions for the exit concentrations against various types of transient inputs, the back transformation is carried out for each of the solutions by means of tables of integral transforms (5) or the Heaviside partial fraction theorem.

This section will be divided into four parts to show how the solutions are obtained for both ideal and non-ideal flow in steady-state and unsteady-state isothermal tubular flow reactors. Only the final results will be given here, and the details of the derivations will be given in Appendix I, II, III, and IV.

Steady-state Non-ideal Flow

The differential equation for the steady-state tubular flow reactor based on the diffusion-type model with axial dispersion and first-order chemical reaction can be obtained from equation (2) by letting the accumulation be zero:

$$0 = \bar{D}\frac{d^2C}{dz^2} - \bar{u}\frac{dC}{dz} - kC \qquad (7)$$

The following transformations of the variables in Equation (7) lead to the dimensionless form of the differential equation:

$$\frac{d^2\gamma}{d\eta^2} - 2M\frac{d\gamma}{d\eta} - 2MR\gamma = 0 \qquad (8)$$

where $\gamma = \frac{C}{C_1}$; $\eta = \frac{z}{L}$; $M = \frac{\bar{u}L}{2\bar{D}}$ and $R = \frac{kL}{\bar{u}}$

The four sets of boundary conditions tabulated in Table 2 of the previous section are transformed likewise and are summarized in Table 3.

Table 3. Transformed forms of boundary conditions.

Boundary Conditions	Inlet ($\eta = 0$)	Outlet ($\eta = 1$)
B.C.(I)	$\frac{d\gamma}{d\eta} = 2M\left[\gamma(0^+) - 1\right]$	$\frac{d\gamma}{d\eta} = 0$
B.C.(II)	$\gamma = 1$	$\frac{d\gamma}{d\eta} = 0$
B.C.(III)	$\gamma = 1$	$\lim_{\eta\to\infty}\gamma = 0$
B.C.(IV)	$\frac{d\gamma}{d\eta} = 2M\left[\gamma(0^+) - 1\right]$	$\lim_{\eta\to\infty}\gamma = 0$

The details of the solutions of equation (2) which satisfy the above boundary conditions (Table 3) will be given in Appendix I. The results obtained for each of the above four boundary conditions are respectively:

$$\gamma_I(\eta) = 2\left\{\frac{(1+\beta)\exp[M(1+\beta) + M(1-\beta)\eta] - (1-\beta)\exp[M(1+\beta)\eta + M(1-\beta)]}{(1+\beta)^2\exp[M(1+\beta)] - (1-\beta)^2\exp[M(1-\beta)]}\right\} \qquad (9)$$

$$\gamma_{II}(\eta) = \frac{(1+\beta)\exp[M(1+\beta) + M(1-\beta)\eta] - (1-\beta)\exp[M(1+\beta)\eta + M(1-\beta)]}{(1+\beta)\exp[M(1+\beta)] - (1-\beta)\exp[M(1-\beta)]} \qquad (10)$$

$$\gamma_{III}(\eta) = \exp[M(1-\beta)\eta] \tag{11}$$

$$\gamma_{IV}(\eta) = \frac{2\exp[M(1-\beta)\eta]}{1+\beta} \tag{12}$$

where $\beta = \sqrt{1 + \frac{2R}{M}}$

Steady-state Ideal Flow

In practice neither plug flow nor completely mixed flow will occur; there always will be some degree of dispersion due to viscous effects and molecular or eddy diffusion. In order to establish the theoretical limitations involved with regard to the degree of mixing, it is necessary to derive the equations for the ideal flow reactor.

For the completely mixed flow reactor, where $D = \infty$ and $M = 0$, and if the effective volume of the reacting mixture, V, and the volumetric flow rate, v, are constant, the material balance is the same as for a single uniformly stirred tank reactor and may be written as:

$$V \frac{dC}{dt} = vC_1 - vC - kC^n \tag{13}$$

where n is the order of reaction.

For the calculation of the steady-state concentration profile and the conversion for the case of a first-order chemical reaction, equation (13) may be simplified.

$$0 = \frac{v}{V} C_1 - \frac{v}{V} C - \frac{k}{V} C \tag{14}$$

Equation (14) can be transformed into a dimensionless form by the substitutions used for (8) to obtain:

$$R\gamma + \gamma - 1 = 0 \tag{15}$$

or

$$\gamma = \frac{1}{1 + R} \tag{16}$$

For the steady-state plug flow reactor, where $D = 0$ and $M = \infty$, the first term in the left-hand side of equation (8) vanishes to give

$$\frac{d\gamma}{d\eta} = -R\gamma \tag{17}$$

The solution of equation (17) is:

$$\gamma(\eta) = e^{-R\eta} \tag{18}$$

Because of the theoretical limits, which were pointed out early in this section, all the concentrations and conversion calculated from equations (9) through (12) have to lie between the results of equations (16) and (18), which give the minimum and maximum values respectively.

Unsteady-state Non-ideal Flow

When the deviation of the concentration of the traceable component from the steady-state value due to a pertubation of inlet concentration, $C_1(t)$, only is considered, the initial condition may be set as $C(z,0) = 0$. The transfer functions, $W(s)$, defined as the ratio between the Laplace transforms of the inlet and the outlet concentrations, using equation (2) for each set of four boundary conditions are shown in equations (19) through (22). The details of the derivations are shown in Appendix II.

$$W(s) = \frac{4\beta'}{(1+\beta')^2 \exp[-M(1-\beta')] - (1-\beta')^2 \exp[-M(1+\beta')]} \tag{19}$$

$$W(s) = \frac{2\beta'}{(1+\beta') \exp[-M(1-\beta')] - (1-\beta') \exp[-M(1+\beta')]} \tag{20}$$

$$W(s) = \exp[M(1-\beta')] \tag{21}$$

$$W(s) = \frac{2\exp[M(1-\beta')]}{1+\beta'} \tag{22}$$

The basic forms of the inlet concentration variations, $C_1(t)$, and their Laplace transforms, $C_1(s)$, for impulse and step input are:

	$C_1(t)$	$C_1(s)$
Impulse	$C_1 u'(t)$	C_1
Step Input	$C_1 u(t)$	C_1/s

The transforms of the exit concentration, $C_2(L,s)$, are then obtained systematically as the products of the transfer functions, $W(s)$, and the $C_1(s)$ as follows:

Impulse	$C_2(s) = C_1 W(s)$
Step Input	$C_2(s) = C_1 W(s)/s$

The inverse transforms of $C_2(s)$ to obtain the response of exit concentration, $C_2(t)$, against the variation of feed concentration, $C_1(t)$, may be carried out systematically by means of the Heaviside partial fraction theorem. The results are tabulated in Table 4 for each of the boundary conditions. The details of the derivations are given in Appendix III.

Table 4. Response of exit concentration from an isothermal tubular flow reactor.

Transient Inputs	B.C.(I)
Impulse	$\frac{C_2 V}{Q} = \sum_{n=1}^{\infty} \frac{2\,\delta_n\,(M \sin \delta_n + \delta_n \cos \delta_n)}{(M^2 + 2M + \delta_n^2)} \times \mathrm{Exp}\left[M - \left(\frac{M^2 + \delta_n^2 + 2MR}{2M}\right)\tau\right]$ (23)
Step Input	$\frac{C_2(t)}{C_1} = A - 4 \sum_{n=1}^{\infty} \frac{M\,\delta_n\,(M \sin \delta_n + \delta_n \cos \delta_n)}{(M^2 + 2M + \delta_n^2)(M^2 + \delta_n^2 + 2MR)} \times \mathrm{Exp}\left[M - \left(\frac{M^2 + \delta_n^2 + 2MR}{2M}\right)\tau\right]$ (27)
Frequency Response	$A.R. = \frac{b\, e^M}{(\alpha_1^2 + \alpha_2^2)^{\frac{1}{2}}}$ $\phi = -\mathrm{Tan}^{-1}\left(\frac{\alpha_2}{\alpha_1}\right) + a$ (31)

Table 4. (Cont.)

Transient Inputs	B.C.(II)

Impulse

$$\frac{C_2 V}{Q} = \sum_{n=1}^{\infty} \frac{-\delta_n'^2}{(M + M^2 + \delta_n'^2)\cos\delta_n'} \times \text{Exp}\left[M - \left(\frac{M^2 + \delta_n'^2 + 2MR}{2M}\right)\tau\right] \qquad (24)$$

Step Input

$$\frac{C_2(t)}{C_1} = B - 2\sum_{n=1}^{\infty} \frac{\delta_n' \sin\delta_n' (M^2 + \delta_n'^2)}{(M^2 + \delta_n'^2 + M)(M^2 + \delta_n'^2 + 2MR)} \times \text{Exp}\left[M - \left(\frac{M^2 + \delta_n'^2 + 2MR}{2M}\right)\tau\right] \qquad (28)$$

Frequency Response

$$A.R. = \frac{b\, e^{M}}{(\alpha_3^2 + \alpha_4^2)^{\frac{1}{2}}}$$

$$\phi = a - \text{Tan}^{-1}\left(\frac{\alpha_4}{\alpha_3}\right) \qquad (32)$$

Table 4. (Cont.)

Transient Inputs	B.C.(III)
Impulse	$\frac{C_2 V}{Q} = \left(\frac{M}{2\pi}\right)^{\frac{1}{2}} (\tau)^{-1.5} \times \mathrm{Exp}\left[M - \frac{1}{2}\left(M\tau + \frac{M}{\tau} + 2R\tau\right)\right]$ (25)
Step Input	$\frac{C_2(t)}{C_1} = \frac{1}{2}\left\{ e^{M+(M^2+2MR)^{\frac{1}{2}}} \times \mathrm{Erfc}\left[\frac{1}{2}\left(\frac{2M}{\tau}\right)^{\frac{1}{2}} + \frac{1}{2}(2M\tau + 4R\tau)^{\frac{1}{2}}\right] + e^{M-(M^2+2MR)^{\frac{1}{2}}} \times \mathrm{Erfc}\left[\frac{1}{2}\left(\frac{2M}{\tau}\right) - \frac{1}{2}(2M\tau + 4R\tau)^{\frac{1}{2}}\right]\right\}$ (29)
Frequency Response	$A.R. = e^{M-x}$ $\phi = -y$ (33)

Table 4. (Cont.)

Transient Inputs	B.C.(IV)

Impulse

$$\frac{C_2 V}{Q} = \left(\frac{2M}{\pi}\right)^{\frac{1}{2}} (\tau)^{-\frac{1}{2}} \times \mathrm{Exp}\left[M - \tfrac{1}{2}\left(M\tau + \frac{M}{\tau} + 2R\tau\right)\right]$$

$$-M\,\mathrm{Exp}(2M - R\tau)\,\mathrm{Erfc}\left[\left(\frac{M}{2\tau}\right)^{\frac{1}{2}} + \left(\frac{M\tau}{2}\right)^{\frac{1}{2}}\right] \quad (26)$$

Step Input

$$\frac{C_2(t)}{C_1} = \int_0^{\tau} \left\{\left(\frac{2M}{\pi}\right)(\lambda)^{-\frac{1}{2}} \mathrm{Exp}\left[M - \tfrac{1}{2}\left(M\lambda + \frac{M}{\lambda} - 2R\lambda\right)\right]\right.$$

$$\left. -M\,\mathrm{Exp}(2M - R\tau)\,\mathrm{Erfc}\left[\left(\frac{M}{2\tau}\right)^{\frac{1}{2}} + \left(\frac{M\tau}{2}\right)^{\frac{1}{2}}\right]\right\} d\lambda$$

(30)

Frequency Response

$$A.R. = \frac{2\, e^{M-x}}{\left[(1 + x/M)^2 + \sin^2 a\right]^{\frac{1}{2}}}$$

$$\phi = -y - \mathrm{Tan}^{-1}\left(\frac{\sin a}{1 + x/M}\right) \quad (34)$$

The amplitude ratios and phase shifts of the frequency response functions caused by a sinusoidal input of amplitude C_1 and angular frequency ω, are obtained by means of a Fourier transformation. Thus, a performance function, defined as the ratio of the Fourier transform of the output to that of the input, is obtained by replacing s by $j\omega$ in equations (19) through (22) for each of the boundary conditions respectively, where $j = \sqrt{-1}$. From these performance functions, the computable forms of the amplitude ratios and phase shifts are obtained by changing into polar coordinates. These are also summarized in Table 4. The details of derivations will be given in Appendix IV.

Unsteady-state Ideal Flow

For a completely mixed reactor, where $D = \infty$ and $M = 0$, the Laplace transform of equation (13) with respect to time with zero initial concentration gives a transfer function of the following form:

$$W(s) = \frac{1}{1 + (s+k)\theta} \tag{35}$$

Substitution of the corresponding transient inputs and their Laplace transforms into equation (35) and inverse transformation of the resulting equations gives:

Impulse
$$\frac{C_2(t)V}{Q} = e^{-(1+R)\tau} \tag{36}$$

Step Input
$$\frac{C_2(t)}{C_1} = \frac{1}{1+R}\left[1-e^{-(1+R)\tau}\right] \tag{37}$$

Frequency Response

$$\left[P.F.\right]_{M=0}(j\omega) = \frac{1}{(1+R)^2 + (\omega\theta)^2}\, e^{-j\tan^{-1}\left(\frac{\omega\theta}{1+R}\right)} \qquad (38)$$

where P.F. = the performance function.

Q = quantity of tracer injected in the impulse function.

V = effective volume of reactor

$\tau = t/\theta$

For the unsteady-state plug flow reactor, where $\overline{D} = 0$ and $M = \infty$, the first term on the right-hand side of equation (2) vanishes, so that

$$\frac{\partial C}{\partial t} = -\overline{u}\,\frac{\partial C}{\partial z} - kC \qquad (39)$$

The Laplace transform of equation (39) with respect to time with zero initial concentration gives a transfer function of the following form:

$$W(s) = e^{-R-s\theta} \qquad (40)$$

Substitution of the corresponding transient inputs and their Laplace transforms into equation (40) and inverse transformation of the resulting equations gives:

$$\text{Impulse}\quad \frac{C_2(t)\,V}{Q} = e^{-R}\,\delta(\tau-\tau_o) \qquad (41)$$

$$\text{Step Input } \frac{C_2(t)}{C_1} = \begin{cases} 0 & 0 < \tau < 1 \\ e^{-R} & \tau > 1 \end{cases} \tag{42}$$

$$\text{Frequency Response } \left[\text{P.F.}\right]_{M=\infty} (j\omega) = e^{-R}\, e^{-j(\omega\theta)} \tag{43}$$

NUMERICAL COMPUTATION

The numerical computations for steady-state solutions represented by equations (9) through (12), (16) and (18) are straight forward and those for the frequency response solutions represented by equations (31) through (34), (38) and (43) are easy to perform by use of a computer, and no elaboration is required. This section is primarily concerned with the computations for unsteady-state transient inputs, impulse and step inputs, represented by equations (23), (24), (27) and (28). All of these four equations are in series form which requires that a tolerable error be established such that the error term of the series does not exceed 10^{-5}.

The first step in calculating numerical values of these equations was to determine the method of convergence of the equations:

$$\cot \delta = \frac{1}{2}\left(\frac{\delta}{M} - \frac{M}{\delta}\right) \qquad \text{for} \quad \delta_n \tag{44}$$

$$\tan \delta' = + \frac{\delta'}{M} \qquad \text{for} \quad \delta_n' \tag{45}$$

For this purpose, the Newton-Raphson iterative formula was used, namely,

$$\begin{aligned} \delta_n^{[q+1]} &= \delta_n^{[q]} - \frac{f(\delta_n)}{f'(\delta_n')} \\ &= \delta_n^{[q]} + \frac{\cot \delta_n^{[q]} - \frac{1}{2}\left(\frac{\delta_n^{[q]}}{M} - \frac{M}{\delta_n^{[q]}}\right)}{1 + \csc^2 \delta_n^{[q]} + \frac{1}{2}\left(\frac{1}{M} + \frac{M}{\delta_n^{[q]2}}\right)} \end{aligned} \tag{46}$$

with a first approximation of the following:

$$\delta_1^{[o]} = 2$$

$$\delta_n^{[o]} = \delta_{n-1}^{[o]} + \pi \qquad n = 2, 3, 4, \ldots\ldots \tag{47}$$

Equation (47) holds for all values of M.

The n-th term of equations (23), (24), (27) and (28) were then calculated with the δ and δ' thus obtained from equations (44) and (45), and the infinite series is truncated after the n-th term such that $\left|\left(\frac{C_2V}{Q}\right)_n \Big/ \sum_{n=1}^{n}\left(\frac{C_2V}{Q}\right)_n\right|$ is smaller than the maximum tolerable error of 10^{-5}.

The block diagrams and computer programs for use with the IBM 650 computers are presented in Table 5 for equations (23), (24) and (44) and in Table 6 for equations (27), (28) and (45) for R = 0 only.

Table 5-a Block diagram for use with IBM 650 for computing equations (23), (24) and (44).

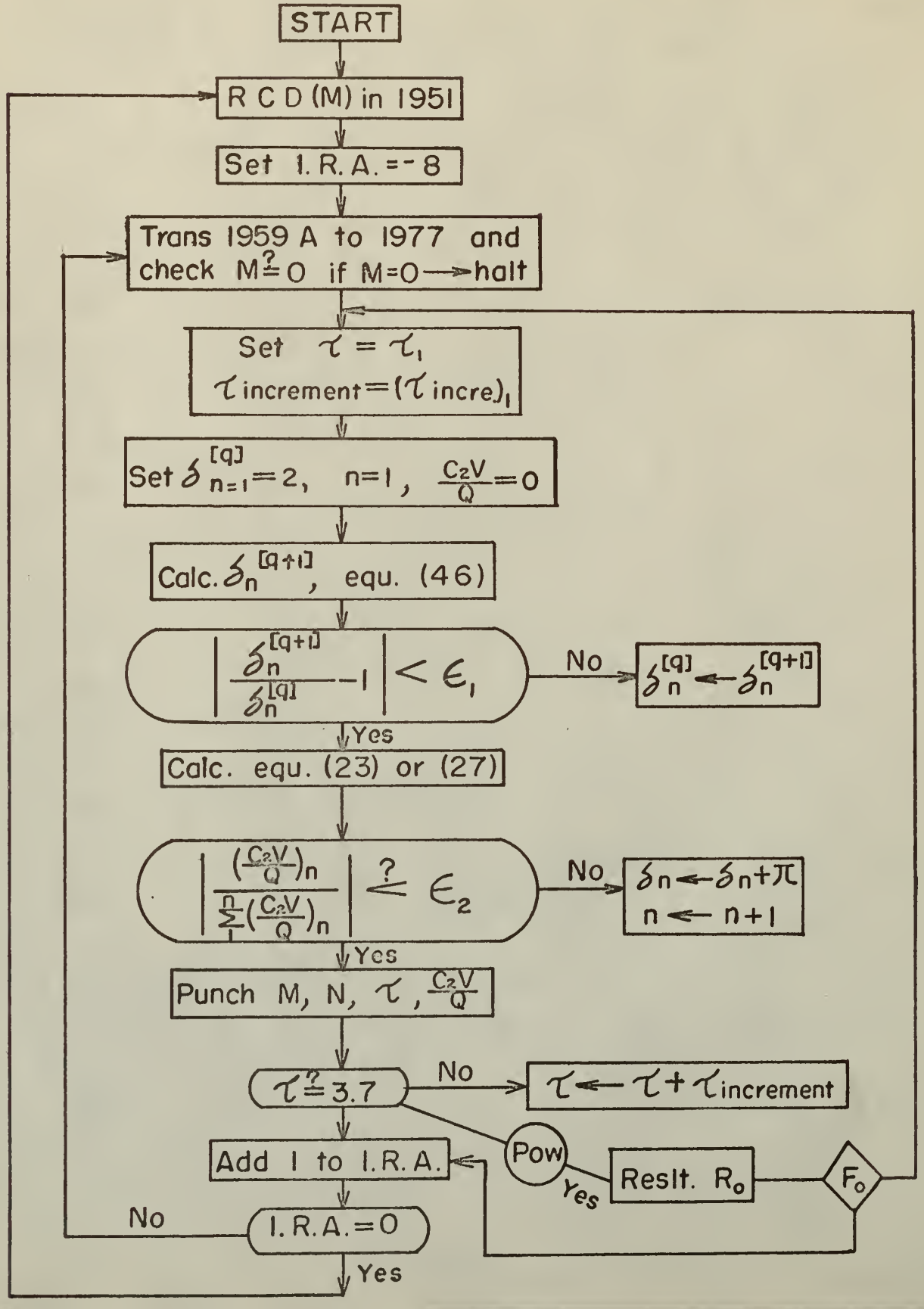

Table 5-b. Computer programs for use with IBM 650 for computing equations (23) (24) and (44)

MODIF	LDD	1582		1572	69	1582	1592
	STD	1570		1592	24	1570	1553
	32	1500	0700	1582	32	1500	0700
	LDD	1563		1553	69	1563	1573
	STD	1571	DEL17	1573	24	1571	0107
1563 = Number of R				1563	00	0006	0000
Program Step No. 202, Connect to 1570				0277	34	0250	1570
	FAD	1500	0700	1570	32	1500	0700
	RAU	1570		1560	60	1570	1580
	AUP	1590		1580	10	1590	1551
	STU	1570		1551	21	1570	1561
	RAU	1571		1561	60	1571	1581
	SUP	1590		1581	11	1590	1552
	STU	1571		1552	21	1571	1562
	NZU	NO	BACK to prog.	1562	44	0030	0777
1590 = Fixed point one				1590	00	0001	0000
	R			1500	00	0000	0000
	R			1501	10	0000	0050
	R			1502	10	0000	0051
	R			1503	50	0000	0051
	R			1504	10	0000	0052
	R			1505	50	0000	0052
	RSA	0008	MODIF	0151	81	0008	1572
Program step No. 255, Connect to 1560				0727	71	1877	1560

```
EXP01   STO  EXP02                                  15  0000  24 0003 0006
        STU  EXP03                                  16  0006  21 0010 0013
        LOO   8007                                  17  0013  69 8007 0019
        STO  EXP04                                  18  0019  24 0022 0025
        RAC   0000                                  19  0025  88 0000 0031
        RAU  EXP03                                  20  0031  60 0010 0015
        BMI  EXP05    EXP06                         21  0015  46 0018 0069
EXP06   FSB  LNTEN                                  22  0069  33 0072 0049
        BMI  EXP07                                  23  0049  46 0002 0053
        AXC   0001    EXP06                         24  0053  58 0001 0069
EXP07   FAD  LNTEN    EXP08                         25  0002  32 0072 0099
EXP05   SXC   0001                                  26  0018  59 0001 0024
        FAD  LNTEN                                  27  0024  32 0072 0149
        BMI  EXP05    EXP08                         28  0149  46 0018 0099
EXP08   STU  EXP03                                  29  0099  21 0010 0063
        STU  EXP09                                  30  0063  21 0068 0021
        RAL  EXP04                                  31  0021  65 0022 0027
        LDO   8007                                  32  0027  69 8007 0033
        STO  EXP04                                  33  0033  24 0022 0075
        RAC   8002                                  34  0075  88 8002 0083
        LDO  FPDNE                                  35  0083  69 0036 0039
        STO  EXP10                                  36  0039  24 0042 0045
        STD  EXP11    EXP12                         37  0045  24 0048 0001
EXP12   RAU  EXP10                                  38  0001  60 0042 0047
        FAO  EXP09                                  39  0047  32 0068 0095
        STU  EXP10                                  40  0095  21 0042 0145
        RAU  EXP11                                  41  0145  60 0048 0103
        FAO  FPDNE                                  42  0103  32 0036 0113
        STU  EXP11                                  43  0113  21 0048 0051
        RAU  EXP09                                  44  0051  60 0068 0023
        FMP  EXP03                                  45  0023  39 0010 0060
        FOV  EXP11                                  46  0060  34 0048 0098
        STU  EXP09                                  47  0098  21 0068 0071
        FOV  EXP10                                  48  0071  34 0042 0092
        FS8  SIZE8                                  49  0092  33 0195 0121
        BMI           EXP12                         50  0121  46 0074 0001
        RAU  EXP10                                  51  0074  60 0042 0097
        AUP  EXP04    EXP02                         52  0097  10 0022 0003
FPONE     10  0000      0051                        53  0036  10 0000 0051
SIZE8     10  0000      0043                        54  0195  10 0000 0043
LNTEN     23  0258      5151                        55  0072  23 0258 5151
EQUCB   STD  CSV02                BEGIN COS         56  0050  24 0153 0056
        NZE  CSV03    CSV04          002  57  0056  45 0110 0011
CSV04   RAU  FPDNE    CSV02          003  58  0011  60 0036 0153
CSV03   BMI  CSV05    CSV06          004  59  0110  46 0163 0014
CSV05   FAO  TWOPI                   005  60  0163  32 0016 0043
        NZE           CSV04          006  61  0043  45 0046 0011
        BMI  CSV05                   007  62  0046  46 0163 0100
        FS8  ONEPI                   008  63  0100  33 0203 0029
        NZE  CSV07    CSV08          009  64  0029  45 0032 0133
CSV08   RSU  FPDNE    CSV02          010  65  0133  61 0036 0153
CSV06   FS8  TWOPI                   011  66  0014  33 0016 0093
        NZE           CSV04          012  67  0093  45 0096 0011
        BMI           CSV06          013  68  0096  46 0199 0014
        FAD  ONEPI                   014  69  0199  32 0203 0079
        NZE  CSV07    CSV08          015  70  0079  45 0032 0133
CSV07   STU  CSV09                   016  71  0032  21 0086 0089
        RSU  FPDNE                   017  72  0089  61 0036 0041
        STU  CSV10                   018  73  0041  21 0146 0249
        STU  CSV11                   019  74  0249  21 0004 0007
        STL  CSV12    CSV13          020  75  0007  20 0061 0064
EQUCA   STO  CSV02                BEGIN SIN         76  0150  24 0153 0106
        NZE           CSV02          021  77  0106  45 0160 0153
        BMI  CSV14    CSV15          022  78  0160  46 0213 0114
CSV14   FAD  TWOPI                   023  79  0213  32 0016 0143
        NZE           CSV02          024  80  0143  45 0196 0153
        BMI  CSV14                   025  81  0196  46 0213 0200
        FS8  ONEPI                   026  82  0200  33 0203 0129
        NZE  CSV16    CSV02          027  83  0129  45 0082 0153
CSV15   FS8  TWOPI                   028  84  0114  33 0016 0193
        NZE           CSV02          029  85  0193  45 0246 0153
        BMI           CSV15          030  86  0246  46 0299 0114
        FAO  ONEPI                   031  87  0299  32 0203 0179
        NZE  CSV16    CSV02          032  88  0179  45 0082 0153
CSV16   STU  CSV09                   033  89  0082  21 0086 0139
        RSU   8003                   034  90  0139  61 8003 0147
        STU  CSV10                   035  91  0147  21 0146 0349
        STU  CSV11                   036  92  0349  21 0004 0057
        LOO  FPONE                   037  93  0057  69 0036 0189
        STD  CSV12    CSV13          038  94  0189  24 0061 0064
CSV13   RAU  CSV12                   039  95  0064  60 0061 0065
        FAD  FPONE                   040  96  0065  32 0036 0263
        STU  CSV17                   041  97  0263  21 0118 0171
        FAO  FPONE                   042  98  0171  32 0036 0313
        STU  CSV12                   043  99  0313  21 0061 0164
        RSU  CSV10                   044 100  0164  61 0146 0101
        FMP  CSV09                   045 101  0101  39 0086 0136
        FMP  CSV09                   046 102  0136  39 0086 0186
        STU  CSV10                   049 105  0111  21 0146 0399
        FDV  CSV11                   050 106  0399  34 0004 0054
        RAM   8003                   051 107  0054  67 8003 0161
        RAU   8002                   052 108  0161  60 8002 0119
        FS8  SIZE8                   053 109  0119  33 0195 0221
        BMI  CSV18                   054 110  0221  46 0124 0125
        RAU  CSV11                   055 111  0125  60 0004 0009
        FAD  CSV10                   056 112  0009  32 0146 0073
        STU  CSV11    CSV13          057 113  0073  21 0004 0064
CSV18   RAU  CSV11    CSV02          058 114  0124  60 0004 0153
FPONE     10  0000      0051        059 115  0036  10 0000 0051
FPTWO     20  0000      0051        060 116  0250  20 0000 0051
TWOPI     62  8318      5351        061 117  0016  62 8318 5351
SIZE8     10  0000      0043        062 118  0195  10 0000 0043
ONEPI     31  4159      2751        063 119  0203  31 4159 2751
XSQOS     50  0000      0049            120  0300  50 0000 0049
FP38      38  0000      0051            121  0350  38 0000 0051
FPD1      10  0000      0051            122  0400  10 0000 0051
SIZEA     10  0000      0045            123  0450  10 0000 0045
SIZE8     10  0000      0044            124  0500  10 0000 0044
EQNO1   LOO  LINKC                       125  1991  69 0044 0197
        STO  LINKA    START              126  0197  24 0550 0253
LINKC   NOP   0000    LINK8              127  0044  00 0000 0148
EQNO8   LOO  LINKD                       128  1992  69 0245 0198
        STO  LINKA    START              129  0198  24 0550 0253
LINKD   FOV  M202     LINK8              130  0245  34 0248 0148
START   RCD   1951                       131  0253  70 1951 0151
        RSA   0008    DEL17              132  0151  81 0008 0107
        FDV  CSV12                 048   104  0168  34 0061 0111
        FDV  CSV17                 047   103  0186  34 0118 0168
DEL17   RAU   1959  A                    133  0107  60 3959 0363
        NZU           DEL18              134  0363  44 0017 0218
        STU   1977                       135  0017  21 1977 0030
        LDO  XSQOS                       136  0030  69 0300 0303
```

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                STO  XINCR                      137   0303   24 0156 0059
                STD  X          DEL15           138   0059   24 1979 0132
DEL15           RSU  FPONE                      139   0132   61 0036 0091
                STU  DELTA                      140   0091   21 0296 0449
                RAB  0000       DEL19           141   0449   82 0000 0005
DEL19           RAC  0000                       142   0005   88 0000 U211
                LDD  1998                       143   0211   69 1998 0201
                STD  N                          144   0201   24 1978 0081
                STD  Y          DEL20           145   0081   24 1980 0183
DEL20           NZB             DEL21           146   0183   42 0236 U037
                AXC  0001       DEL22           147   0236   58 0001 0142
DEL21           RAU  DELTA                      148   0037   60 0296 0251
                FAD  DNEPI                      149   0251   32 0203 0229
                STU  DELTA      DEL06           150   0229   21 0296 0499
DEL06           RAU  DELTA                      151   0499   60 0296 0301
                LDD             E00CA           152   0301   69 0104 0150
                STU  SIN                        153   0104   21 0008 0261
                RAU  DELTA                      154   0261   60 0296 0351
                LDD             E00C8           155   0351   69 0154 0050
                STU  COS                        156   0154   21 0058 0311
                FDV  SIN                        157   0311   34 0008 0108
                STU  COT                        158   0100   21 0012 0115
                FMP  8003                       159   0115   39 8003 0169
                STU  CQTSQ                      160   0169   21 0174 0077
                RAU  FPONE                      161   0077   60 0036 0141
                FDV  M                          162   0141   34 1977 0127
                STU  DEL01                      163   0127   21 0182 0035
                RAU  M                          164   0035   60 1977 0131
                FDV  DELTA                      165   0131   34 0296 0346
                FDV  DELTA                      166   0346   34 0296 0396
                FAD  DEL01                      167   0396   32 0182 0109
                FDV  FPTWO                      168   0109   34 0250 0600
                FAD  CQTSQ                      169   0600   32 0174 0401
                FAD  FPONE                      170   0401   32 0036 0413
                STU  DEL02                      171   0413   21 0268 0271
                RAU  DELTA                      172   0271   60 0296 0451
                FDV  M                          173   0451   34 1977 0177
                STU  DEL03                      174   0177   21 0232 0085
                RAU  M                          175   0085   60 1977 0181
                FDV  DELTA                      176   0181   34 0296 0446
                FSB  DEL03                      177   0446   33 0232 0159
                FDV  FPTWO                      178   0159   34 0250 0650
                FAD  CQT                        179   0650   32 0012 0239
                FDV  DEL02                      180   0239   34 0268 0318
                FAD  DELTA                      181   0318   32 0296 0123
                STU  DEL04                      182   0123   21 0028 0231
                FDV  DELTA                      183   0231   34 0296 0496
                FSB  FPONE                      184   0496   33 0036 0463
                RAM  8003                       185   0463   67 8003 0321
                RAU  8002                       186   0321   60 8002 0279
                FSB  SIZEA                      187   0279   33 0450 0227
                BMI  DEL05                      188   0227   46 0080 0281
                LDD  DEL04                      189   0281   69 0028 0331
                STO  DELTA      DEL06           190   0331   24 0296 0499
DEL05           AXC  0001                       191   0080   58 0001 0286
                LDD  DEL04                      192   0286   69 0028 0381
                STD  DELTA                      193   0381   24 0296 0549
                RAU  DELTA                      194   0549   60 0296 0501
                FMP  8003                       195   0501   39 8003 0055
                STU  DEL07                      196   0055   21 0210 0513
                RAU  M                          197   0513   60 1977 0431
                FMP  8003                       198   0431   39 8003 0135
                FAD  DEL07                      199   0135   32 0210 0087
                STU  TEMPX                      200   0087   21 0192 0295
                FDV  M                          201   0295   34 1977 0277
                FDV  FPTWO                      202   0277   34 0250 0700
                STU  1200  C                    203   0700   21 7200 0353
                RAU  M                          204   0353   60 1977 0481
                FMP  FPTWO                      205   0481   39 0250 0750
                FAD  TEMPX                      206   0750   32 0192 0219
                STU  DEL10                      207   0219   21 0224 0327
                RAU  COS                        208   0327   60 0058 0563
                FMP  DELTA                      209   0563   39 0296 0546
                STU  DEL11                      210   0546   21 0800 0403
                RAU  SIN                        211   0403   60 0008 0613
                FMP  M                          212   0613   39 1977 0377
                FAD  DEL11                      213   0377   32 0800 0427
                FMP  DELTA                      214   0427   39 0296 0596
                FMP  FPTWO                      215   0596   39 0250 U850
                FDV  DEL10                      216   0850   34 0224 U274
                STO  1000  C    DEL22           217   0274   21 7000 0142
DEL22           LDD  1200  C                    218   0142   69 7200 0453
                STD  M202                       219   0453   24 0248 0551
                LDD  1000  C                    220   0551   69 7000 0503
                STD  NEXTM                      221   0503   24 0206 0209
                RAU  N                          222   0209   60 1978 0233
                FAD  FPONE                      223   0233   32 0036 0663
                STU  N                          224   0663   21 1978 0531
                RAU  M202                       225   0531   60 0248 0553
                FMP  X                          226   0553   39 1979 0329
                STU  DEL09                      227   0329   21 0034 0137
                RAU  M                          228   0137   60 1977 0581
                FSB  DEL09                      229   0581   33 0034 0361
                LDD             EXP01           230   0361   69 0214 0000
                FMP  NEXTM      LINKA           231   0214   39 0206 0550
LINKB           STU  DEL12                      232   0148   21 0052 0105
                FAD  Y                          233   0105   32 1980 0157
                STU  Y                          234   0157   21 1980 0283
                RAO  DEL12                      235   0283   60 0052 0207
                FDV  Y                          236   0207   34 1980 0130
                RAM  8003                       237   0130   67 8003 0187
                RAU  8002                       238   0187   60 8002 0345
                FSB  SIZEB                      239   0345   33 050U 0477
                BMI  DEL13      DEL20           240   0477   46 0180 U183
DEL13           PCH  1977                       241   0180   71 1977 0527
                RAU  X                          242   0527   60 1979 U333
                FAO  XINCR                      243   0333   32 0156 0383
                STU  X                          244   0383   21 1979 0282
                AXB  0001                       245   0282   52 0001 0038
                FSB  FP38                       246   0038   33 0350 0577
                NZU             DEL14           247   0577   44 0631 0332
                FAD  FP38                       248   0631   32 0350 0627
                FSB  FP01                       249   0627   33 0400 0677
                NZU  DEL19      DEL16           250   0677   44 0005 0382
DEL16           RAO  XINCR                      251   0382   60 0156 0411
                FMP  FPTWO                      252   0411   39 0250 0900
                STO  XINCR      DEL19           253   0900   21 0156 0005
DEL14           PCH  1877                       254   0332   71 1877 0727
                PCH  1877                       255   0727   71 1877 U777
                AXA  0001                       256   0777   50 0001 0433
                NZA  DEL17      START           257   0433   40 0107 0253
DEL18           HLT  9999       9999            258   0218   01 9999 9999
```

Table 6 – a. Block diagram for use with IBM 650 for computing equations (27) and (28).

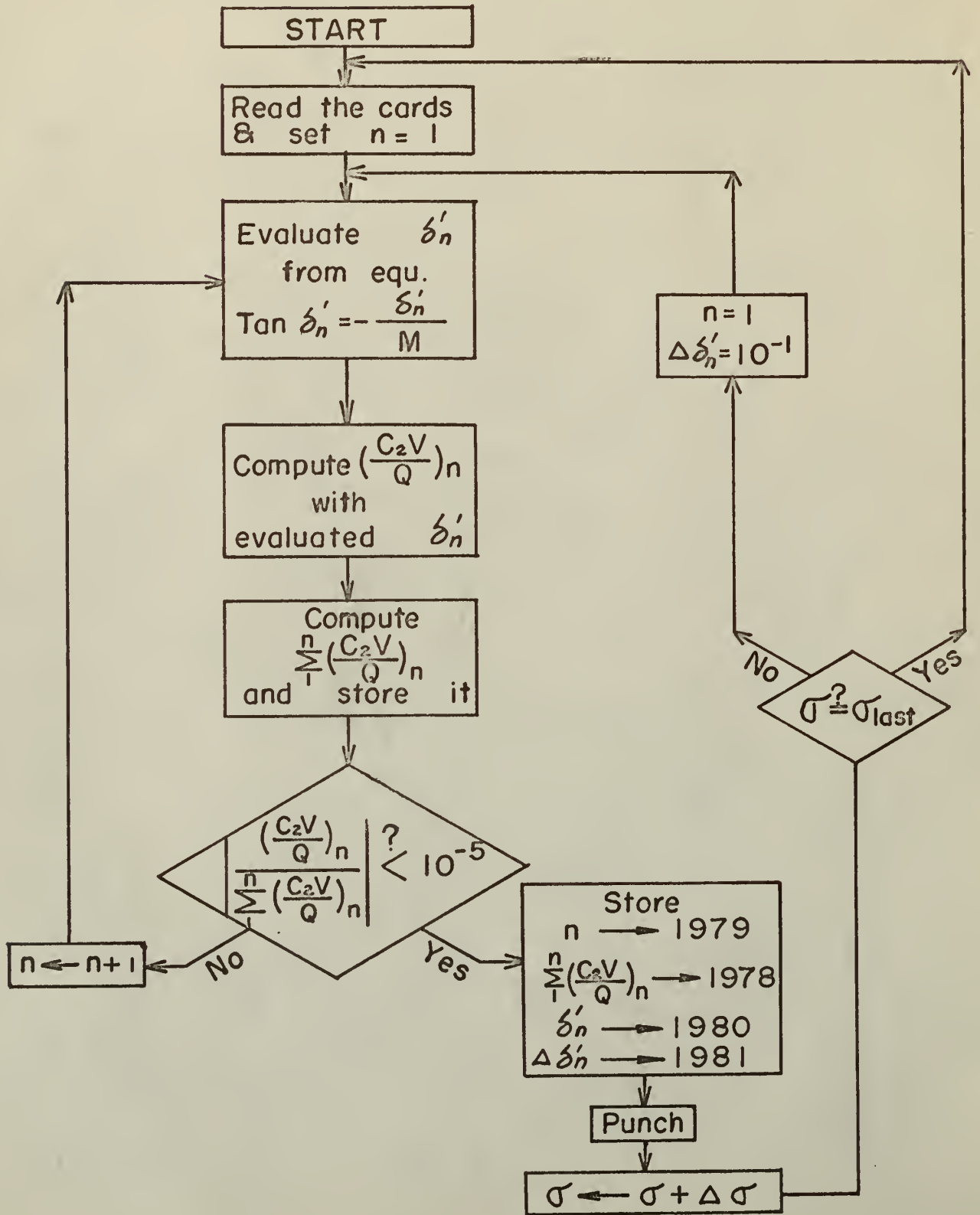

Table 6—b. Block diagram for use with IBM 650 for computing equation (45).

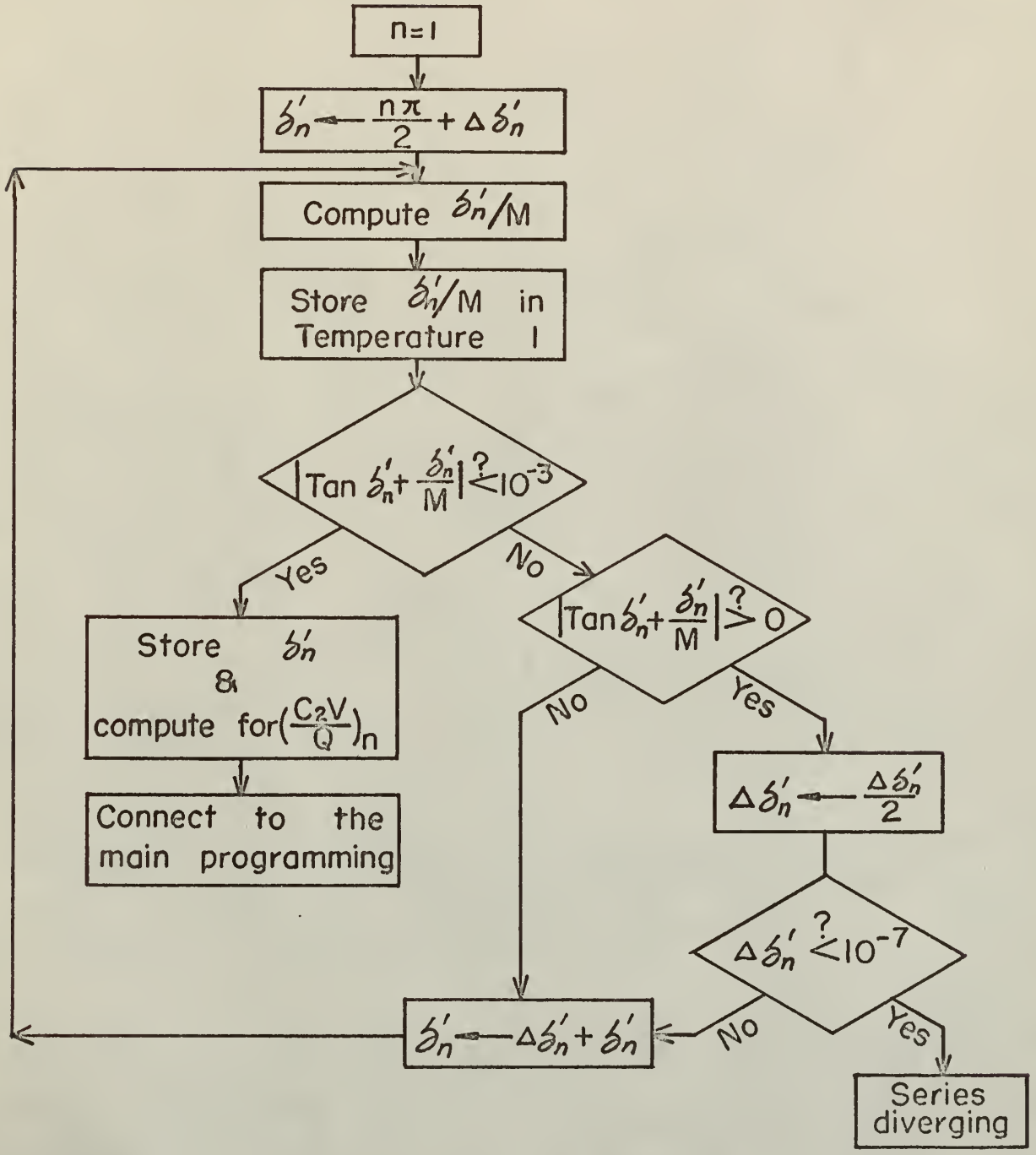

Table 6 – c. Computer program for use with IBM 650 for computing equations (27) & (45)

```
                BLR   1000    1999                1    0000    00 0000 0000
PRES1            10   0000    0048                2    0000    10 0000 0048
INCOT            10   0000    0050                3    0050    10 0000 0050
DEC01            10   0000    0050                4    0100    10 0000 0050
HAFPI            15   7079    6351                5    0150    15 7079 6351
FP043            10   0000    0043                6    0200    10 0000 0043
ACUCY            10   0000    0044                7    0250    10 0000 0044 ——0300 50 0000 0050
MXTAU            36   0000    0051                9    0350    36 0000 0051
D TAU            50   0000    0049               10    0400    50 0000 0049
DIVGE            99   9999    9999               11    0450    99 9999 9999
ONE 5            10   5000    0051               12    0500    10 5000 0051
FP 3             30   0000    0051               13    0550    30 0000 0051
START           RCO   1951                       14    0600    70 1951 0001
                RAU   1STAU   SET                15    0001    60 0300 0005
SET             STU   1977                       16    0005    21 1977 0030
                LDO   DEC01                      17    0030    69 0100 0003
                STD   INCOT                      18    0003    24 0050 0053
                LOD   1951                       19    0053    69 1951 0004
                STO   1984                       20    0004    24 1984 0037
                LOO   FPONE                      21    0037    69 0040 0043
                STD   1979    AKIOO              22    0043    24 1979 0032
AKIOO           RAU   1979                       23    0032    60 1979 0033
                FMP   ONEPI                      24    0033    39 0036 0086
                STU   N PI                       25    0086    21 0090 0093
                RAU   1979                       26    0093    60 1979 0083
                FMP   FPTWO                      27    0083    39 0136 0186
                FSB   FPONE                      28    0186    33 0040 0017
                FMP   HAFPI                      29    0017    39 0150 0650
                FAO   INCOT                      30    0650    32 0050 0027
                FSB   N PI                       31    0027    33 0090 0067
                NZU             RETUN            32    0067    44 0021 0022
                BMI             RETUN            33    0021    46 0024 0022
                FAO   N PI                       34    0024    32 0090 0117
                STU   LOCTO     COMPU            35    0117    21 0072 0025
COMPU           FDV   1951                       36    0025    34 1951 0051
                STU   TEMP1                      37    0051    21 0006 0009
                RAU   LOCTO                      38    0009    60 0072 0077
                LOD             EOOC8            39    0077    69 0080 0133
                STU   COSIN                      40    0080    21 0034 0087
                RAU   LOCTO                      41    0087    60 0072 0127
                LDO             EOOCA            42    0127    69 0130 0183
                FOV   COSIN                      43    0130    34 0034 0084
                FAO   TEMP1                      44    0084    32 0006 0233
                BMI   ASKNG     CHECK            45    0233    46 0236 0137
ASKNG           FAD   PRES1                      46    0236    32 0000 0177
                NZU   NOSIR     OK               47    0177    44 0031 0082
NOSIR           BMI   MODFY     OK               48    0031    46 0134 0082
MDOFY           RAU   LOCTO                      49    0134    60 0072 0227
                FAD   INCOT                      50    0227    32 0050 0277
                STU   LOCTO     COMPU            51    0277    21 0072 0025
CHECK           FSB   PRES1                      52    0137    33 0000 0327
                NZU   TESTQ     OK               53    0327    44 0081 0082
TESTQ           BMI   OK        RETUN            54    0081    46 0082 0022
RETUN           RAU   INCOT                      55    0022    60 0050 0055
                FOV   FPTWO                      56    0055    34 0136 0286
                FSB   FP043                      57    0286    33 0200 0377
                BMI   MINUS                      58    0377    46 0180 0131
                FAO   FP043                      59    0131    32 0200 0427
                STU   INCDI                      60    0427    21 0050 0103
                RAU   LOCTD                      61    0103    60 0072 0477
                FSB   INCDT                      62    0477    33 0050 0527
                STU   LOCTO     COMPU            63    0527    21 0072 0025
OK              RAU   LOCTO                      64    0082    60 0072 0577
                FMP   LOCTO                      65    0577    39 0072 0122
                STU   SQR D                      66    0122    21 0026 0029
                RAU   1951                       67    0029    60 1951 0105
                FMP   1951                       68    0105    39 1951 0101
                STU   MSQRE                      69    0101    21 0056 0059
                FAO   SQR D                      70    0059    32 0026 0153
                FDV   1951                       71    0153    34 1951 0151
                FOV   FPTWO                      72    0151    34 0136 0336
                FMP   1977                       73    0336    39 1977 0627
                STU   BOX 2                      74    0627    21 0132 0035
                RAU   1951                       75    0035    60 1951 0155
                FSB   BOX 2                      76    0155    33 0132 0109
                LDO             EOOLR            77    0109    69 0012 0015
                STU   1982                       78    0012    21 1982 0085
                RAU   1951                       79    0085    60 1951 0205
                FAD   MSQRE                      80    0205    32 0056 0283
                FAO   SQR 0                      81    0283    32 0026 0203
                FMP   COSIN                      82    0203    39 0034 0184
                STU   BOX1                       83    0184    21 0038 0041
                RSU   SQR D                      84    0041    61 0026 0181
                FDV   BOX1                       85    0181    34 0038 0088
                FMP   1982                       86    0088    39 1982 0182
                STU   1981                       87    0182    21 1981 0234
                FAO   1978                       88    0234    32 1978 0255
                STU   1978                       89    0255    21 1978 0231
                RAU   1981                       90    0231    60 1981 0135
                FOV   1978                       91    0135    34 1978 0028
                RAM   8003                       92    0028    67 8003 0185
                RAU   8002                       93    0185    60 8002 0143
                FSB   ACUCY                      94    0143    33 0250 0677
                BMI   THANK                      95    0677    46 0230 0281
                RAU   1979                       96    0281    60 1979 0333
                FAO   FPONE                      97    0333    32 0040 0167
                STU   1979                       98    0167    21 1979 0232
                LDO   DEC01                      99    0232    69 0100 0253
                STO   INCOT     AKIOO           100    0253    24 0050 0032
THANK           LDO   LOCTO                     101    0230    69 0072 0075
                STO   1980                      102    0075    24 1980 0383
                LOO   COSIN                     103    0383    69 0034 0187
                STO   1983                      104    0187    24 1983 0386
                PCH   1977      AOTAU           105    0386    71 1977 0727
AOTAU           LDD   1999                      106    0727    69 1999 0002
                STD   1978                      107    0002    24 1978 0331
                RAU   1977                      108    0331    60 1977 0381
```

```
                FAD   D TAU                                  109   0381   32 0400 0777
                FSB   ONE 5                                  110   0777   33 0500 0827
                BMI                GD1                       111   0827   46 0280 0431
                FAD   ONE 5        SET                       112   0280   32 0500 0005
GD1             FAB   ONE 5                                  113   0431   32 0500 0877
                FAD   D TAU                                  114   0877   32 0400 0927
                FSB   MXTAU                                  115   0927   33 0350 0977
                BMI                CONN1                     116   0977   46 0330 0481
                FAD   MXTAU        SET                       117   0330   32 0350 0005
CONN1           LOD   DECO1                                  118   0481   69 0100 0303
                STD   INCDT        START                     119   0303   24 0050 0600
MINUS           LDD   DIVGE                                  120   0180   69 0450 0353
                STD    1983        PUNCH                     121   0353   24 1983 0436
PUNCH           PCH    1977                                  122   0436   71 1977 0078
                LDD    1999                                  123   0070   69 1999 0052
                STD    1983        AOTAU                     124   0052   24 1983 0727
EOOC8           STD   CSV02                    BEGIN CDS     125   0133   24 0486 0039
                NZE   CSV03        CSV04          002        126   0039   45 0042 0193
CSV04           RAU   FPONE        CSV02          003        127   0193   60 0040 0486
CSV03           BMI   CSV05        CSV06          004        128   0042   46 0045 0046
CSV05           FAD   TWDPI                        005        129   0045   32 0048 0125
                NZE                CSV04          006        130   0125   45 0128 0193
                BMI   CSV05                       007        131   0128   46 0045 0282
                FSB   ONEPI                       008        132   0282   33 0036 0013
                NZE   CSV07        CSV08          009        133   0013   45 0016 0217
CSV08           RSU   FPONE        CSV02          010        134   0217   61 0040 0486
CSV06           FSB   TWDPI                        011        135   0046   33 0048 0175
                NZE                CSV04          012        136   0175   45 0178 0193
                BMI                CSV06          013        137   0178   46 0531 0046
                FAD   ONEPI                       014        138   0531   32 0036 0063
                NZE   CSV07        CSV08          015        139   0063   45 0016 0217
CSV07           STU   CSV09                        016        140   0016   21 0020 0023
                RSU   FPONE                       017        141   0023   61 0040 0095
                STU   CSV10                       018        142   0095   21 0700 0403
                STU   CSV11                       019        143   0403   21 0008 0011
                STL   CSV12        CSV13          020        144   0011   20 0065 0018
EOOCA           STD   CSV02                    BEGIN SIN     145   0183   24 0486 0089
                NZE                CSV02          021        146   0089   45 0092 0486
                BMI   CSV14        CSV15          022        147   0092   46 0145 0096
CSV14           FAD   TWDPI                        023        148   0145   32 0048 0225
                NZE                CSV02          024        149   0225   45 0228 0486
                BMI   CSV14                       025        150   0228   46 0145 0332
                FSB   ONEPI                       026        151   0332   33 0036 0113
                NZE   CSV16        CSV02          027        152   0113   45 0066 0486
CSV15           FSB   TWDPI                        028        153   0096   33 0048 0275
                NZE                CSV02          029        154   0275   45 0278 0486
                BMI                CSV15          030        155   0278   46 0581 0096
                FAD   ONEPI                       031        156   0581   32 0036 0163
                NZE   CSV16        CSV02          032        157   0163   45 0066 0486
CSV16           STU   CSV09                        033        158   0066   21 0020 0073
                RSU    8003                        034        159   0073   61 8003 0631
                STU   CSV10                       035        160   0631   21 0700 0453
                STU   CSV11                       036        161   0453   21 0008 0061
                LDD   FPONE                       037        162   0061   69 0040 0243
                STD   CSV12        CSV13          038        163   0243   24 0065 0018
CSV13           RAU   CSV12                       039        164   0018   60 0065 0019
                FAD   FPONE                       040        165   0019   32 0040 0267
                STU   CSV17                       041        166   0267   21 0172 0325
                FAD   FPONE                       042        167   0325   32 0040 0317
                STU   CSV12                       043        168   0317   21 0065 0068
                RSU   CSV10                       044        169   0068   61 0700 0305
                FMP   CSV09                       045        170   0305   39 0020 0070
                FMP   CSV09                       046        171   0070   39 0020 0120
                FDV   CSV17                       047        172   0120   34 0172 0222
                FDV   CSV12                       048        173   0222   34 0065 0115
                STU   CSV10                       049        174   0115   21 0700 0503
                FDV   CSV11                       050        175   0503   34 0008 0058
                RAM    8003                       051        176   0058   67 8003 0165
                RAU    8002                       052        177   0165   60 8002 0123
                FSB   SIZE8                       053        178   0123   33 0076 0553
                BMI   CSV18                       054        179   0553   46 0106 0007
                RAU   CSV11                       055        180   0007   60 0008 0213
                FAD   CSV10                       056        181   0213   32 0700 0328
                STU   CSV11        CSV13          057        182   0328   21 0008 0018
CSV18           RAU   CSV11        CSV02          058        183   0106   60 0008 0486
FPONE            10    0000         0051          059        184   0040   10 0000 0051
FPTWD            20    0000         0051          060        185   0136   20 0000 0051
TWOPI            62    8318         5351          061        186   0048   62 8318 5351
SIZE8            10    0000         0043          062        187   0076   10 0000 0043
ONEPI            31    4159         2751          063        188   0036   31 4159 2751
EOOLR           STO   EXP02                        001        189   0015   24 0118 0071
                STU   EXP03                        002        190   0071   21 0126 0079
                LDD    8007                        003        191   0079   69 8007 0235
                STD   EXP04                        004        192   0235   24 0138 0091
                RAC    0000                        005        193   0091   88 0000 0047
                RAU   EXP03                        006        194   0047   60 0126 0681
                BMI   EXP05        EXP06          007        195   0681   46 0284 0285
EXP06           FSB   LNTEN                        008        196   0285   33 0188 0215
                BMI   EXP07                        009        197   0215   46 0168 0069
                AXC    0001        EXP06          010        198   0069   58 0001 0285
EXP07           FAD   LNTEN        EXP08          011        199   0168   32 0188 0265
EXP05           SXC    0001                        012        200   0284   59 0001 0140
                FAD   LNTEN                        013        201   0140   32 0188 0315
                BMI   EXP05        EXP08          014        202   0315   46 0284 0265
EXP08           STU   EXP03                        015        203   0265   21 0126 0129
                STU   EXP09                        016        204   0129   21 0334 0237
                RAL   EXP04                        017        205   0237   65 0138 0293
                LOD    8007                        018        206   0293   69 8007 0049
                STD   EXP04                        019        207   0049   24 0138 0141
                RAC    8002                        020        208   0141   88 8002 0099
                LDD   FPONE                        021        209   0099   69 0040 0343
                STD   EXP10                        022        210   0343   24 0146 0149
                STD   EXP11        EXP12          023        211   0149   24 0102 0355
EXP12           RAU   EXP10                        024        212   0355   60 0146 0201
                FAD   EXP09                        025        213   0201   32 0334 0111
                STU   EXP10                        026        214   0111   21 0146 0199
                RAU   EXP11                        027        215   0199   60 0102 0057
                FAD   FPONE                        028        216   0057   32 0040 0367
                STU   EXP11                        029        217   0367   21 0102 0405
                RAU   EXP09                        030        218   0405   60 0334 0139
                FMP   EXP03                        031        219   0139   39 0126 0176
                FDV   EXP11                        032        220   0176   34 0102 0152
                STU   EXP09                        033        221   0152   21 0334 0287
                FDV   EXP10                        034        222   0287   34 0146 0196
                FSB   SIZE8                        035        223   0196   33 0076 0603
                BMI                EXP12           036        224   0603   46 0156 0355
                RAU   EXP10                        037        225   0156   60 0146 0251
                AUP   EXP04        EXP02           038        226   0251   10 0138 0119
FPONE            10    0000         0051          039        227   0040   10 0000 0051
SIZE8            10    0000         0043          040        228   0076   10 0000 0043
LNTEN            23    0258         5151          041        229   0188   23 0258 5151
```

Table 6-d Computer program for use with IBM 650 for computing equations (28) & (45).

```
          BLR   1000    1999                1    0000   00 0000 0000
PRES1      10   0000    0048                2    0000   10 0000 0048
INCDT      10   0000    0050                3    0050   10 0000 0050
DEC01      10   0000    0050                4    0100   10 0000 0050
HAFPI      15   7079    6351                5    0150   15 7079 6351
FP043      10   0000    0043                6    0200   10 0000 0043
ACUCY      10   0000    0044                7    0250   10 0000 0044
1STAU      50   0000    0049                8    0300   50 0000 0049
MXTAU      36   0000    0051                9    0350   36 0000 0051
D TAU      50   0000    0049               10    0400   50 0000 0049
DIVGE      99   9999    99.99              11    0450   99 9999 9999
ONE 5      10   5000    0051               12    0500   10 5000 0051
FP 3       30   0000    0051               13    0550   30 0000 0051
START      RCD  1951                       14    0600   70 1951 0001
           RAU  1STAU   SET                15    0001   60 0300 0005
SET        STU  1977                       16    0005   21 1977 0030
           LOD  DEC01                      17    0030   69 0100 0003
           STD  INCDT                      18    0003   24 0050 0053
           LOO  1951                       19    0053   69 1951 0004
           STO  1984                       20    0004   24 1984 0037
           LOD  FPONE                      21    0037   69 0040 0043
           STD  1979    AKIOO              22    0043   24 1979 0032
AKIOO      RAU  1979                       23    0032   60 1979 0033
           FMP  ONEPI                      24    0033   39 0036 0086
           STU  N PI                       25    0086   21 0090 0093
           RAU  1979                       26    0093   60 1979 0083
           FMP  FPTWO                      27    0083   39 0136 0186
           FS8  FPONE                      28    0186   33 0040 0017
           FMP  HAFPI                      29    0017   39 0150 0650
           FAO  INCOT                      30    0650   32 0050 0027
           FS8  N PI                       31    0027   33 0090 0067
           NZU          RETUN              32    0067   44 0021 0022
           BWI          RETUN              33    0021   46 0024 0022
           FAD  N PI                       34    0024   32 0090 0117
           STU  LOCTD   COMPU              35    0117   21 0072 0025
COMPU      FDV   1951                      36    0025   34 1951 0051
           STU  TEMP1                      37    0051   21 0006 0009
           RAU  LOCTD                      38    0009   60 0072 0077
           LDO          EOOCB              39    0077   69 0080 0133
           STU  COSIN                      40    0080   21 0034 0087
           RAU  LOCTD                      41    0087   60 0072 0127
           LDD          EOOCA              42    0127   69 0130 0183
                                           43    0130   21 0039 0137
           FDV  COSIN                      44    0137   34 0034 0134
           FAD  TEMP1                      45    0134   32 0006 0233
           BWI  ASKNG   CHECK              46    0233   46 0236 0187
ASKNG      FAD  PRES1                      47    0236   32 0000 0177
           NZU  NOSIR   OK                 48    0177   44 0031 0082
NOSIR      BWI  MODFY   OK                 49    0031   46 0184 0082
MODFY      RAU  LOCTO                      50    0184   60 0072 0227
           FAD  INCDT                      51    0227   32 0050 0277
           STU  LOCTO   COMPU              52    0277   21 0072 0025
CHECK      FS8  PRES1                      53    0187   33 0000 0327
           NZU  TESTQ   OK                 54    0327   44 0081 0082
TESTQ      BWI  OK      RETUN              55    0081   46 0082 0022
RETUN      RAU  INCDT                      56    0022   60 0050 0055
           FDV  FPTWO                      57    0055   34 0136 0286
           FS8  FP043                      58    0286   33 0200 0377
           BWI  MINUS                      59    0377   46 0180 0131
           FAD  FP043                      60    0131   32 0200 0427
           STU  INCOT                      61    0427   21 0050 0103
           RAU  LOCTD                      62    0103   60 0072 0477
           FS8  INCDT                      63    0477   33 0050 0527
           STU  LOCTD   COMPU              64    0527   21 0072 0025
OK         RAU  LOCTD                      65    0082   60 0072 0577
           FMP  LOCTD                      66    0577   39 0072 0122
           STU  SQR D                      67    0122   21 0026 0029
           RAU   1951                      68    0029   60 1951 0105
                                           69    0105   39 1951 0155
           STU  MSQRE                      70    0155   21 0010 0013
           FAO  SQR D                      71    0013   32 0026 0153
           FDV   1951                      72    0153   34 1951 0101
           FDV  FPTWO                      73    0101   34 0136 0336
           FMP   1977                      74    0336   39 1977 0627
           STU  BOX 2                      75    0627   21 0132 0035
           RAU   1951                      76    0035   60 1951 0205
           FS8  BOX 2                      77    0205   33 0132 0059
           LDD          EOOLR              78    0059   69 0012 0015
           STU   1982                      79    0012   21 1982 0085
           RAU   1951                      80    0085   60 1951 0255
           FAD  MSQRE                      81    0255   32 0010 0237
           FAD  SQR D                      82    0237   32 0026 0203
           STU  BOX 3                      83    0203   21 0008 0011
           RAU  LOCTD                      84    0011   60 0072 0677
           FMP  FPTWO                      85    0677   39 0136 0386
           FMP  SIGN                       86    0386   39 0039 0089
           FDV  BOX 3                      87    0089   34 0008 0058
           FMP   1982                      88    0058   39 1982 0182
           STU   1981                      89    0182   21 1981 0234
           FAO  SUM                        90    0234   32 0287 0063
           STU  SUM                        91    0063   21 0287 0140
           RAU   1981                      92    0140   60 1981 0135
           FDV  SUM                        93    0135   34 0287 0337
           RAM   8003                      94    0337   67 8003 0045
           RAU   8002                      95    0045   60 8002 0253
           FS8  ACUCY                      96    0253   33 0250 0727
           BWI  THANK                      97    0727   46 0230 0181
           RAU   1979                      98    0181   60 1979 0283
           FAO  FPONE                      99    0283   32 0040 0167
           STU   1979                     100    0167   21 1979 0232
           LDO  DEC01                     101    0232   69 0100 0303
           STO  INCDT   AKIOO             102    0303   24 0050 0032
THANK      LDD  LOCTD                     103    0230   69 0072 0075
           STD   1980                     104    0075   24 1980 0333
```

```
                LOO   SUGN                                  105   0333   69 0084 0387
                STO    1983                                 106   0387   24 1983 0436
                RAU   FPONE                                 107   0436   60 0040 0095
                FSB   SUM                                   108   0095   33 0287 0113
                STU    1978                                 109   0113   21 1978 0231
                PCH    1977    AOTAU                        110   0231   71 1977 0777
AOTAU           LOO    1999                                 111   0777   69 1999 0002
                STO   SUM                                   112   0002   24 0287 0190
                RAU    1977                                 113   0190   60 1977 0281
                FAD   D TAU                                 114   0281   32 0400 0827
                FSB   ONE 5                                 115   0827   33 0500 0877
                BMI           GO1                           116   0877   46 0280 0331
                FAD   ONE 5   SET                           117   0280   32 0500 0005
GO1             FAD   ONE 5                                 118   0331   32 0500 0927
                FAD   D TAU                                 119   0927   32 0400 0977
                FSB   MXTAU                                 120   0977   33 0350 0028
                BMI           CONN1                         121   0028   46 0381 0282
                FAD   MXTAU   SET                           122   0381   32 0350 0005
CONN1           LOD   DECO1                                 123   0282   69 0100 0353
                STO   INCDT   START                         124   0353   24 0050 0600
MINUS           LOD   DIVGE                                 125   0180   69 0450 0403
                STO    1983   PUNCH                        126   0403   24 1983 0486
PUNCH           PCH    1977                                 127   0486   71 1977 0078
                LOD    1999                                 128   0078   69 1999 0052
                STO    1983   AOTAU                        129   0052   24 1983 0777
EOOC8           STO   CSV02          BEGIN  COS    002     130   0133   24 0536 0139
                NZE   CSV03   CSV04                 002     131   0139   45 0042 0143
CSV04           RAU   FPONE   CSV02                 003     132   0143   60 0040 0536
CSV03           BMI   CSV05   CSV06                 004     133   0042   46 0145 0046
CSV05           FAD   TWOPI                         005     134   0145   32 0048 0125
                NZE           CSV04                 006     135   0125   45 0128 0143
                BMI   CSV05                         007     136   0128   46 0145 0332
                FSB   ONEPI                         008     137   0332   33 0036 0163
                NZE   CSV07   CSV08                 009     138   0163   45 0016 0217
CSV08           RSU   FPONE   CSV02                 010     139   0217   61 0040 0536
CSV06           FSB   TWOPI                         011     140   0046   33 0048 0175
                NZE           CSV04                 012     141   0175   45 0178 0143
                BMI           CSV06                 013     142   0178   46 0431 0046
                FAD   ONEPI                         014     143   0431   32 0036 0213
                NZE   CSV07   CSV0H                 015     144   0213   45 0016 0217
CSV07           STU   CSV09                         016     145   0016   21 002U 0023
                RSU   FPONE                         017     146   0023   61 0040 0195
                STU   CSV10                         018     147   0195   21 0750 0453
                STU   CSV11                         019     148   0453   21 0108 0061
                STL   CSV12   CSV13                 020     149   0061   20 0065 0018
EOOCA           STO   CSV02          BEGIN  SIN    021     150   0183   24 0536 0189
                NZE           CSV02                 021     151   0189   45 0092 0536
                BMI   CSV14   CSV15                 022     152   0092   46 0245 0096
CSV14           FAD   TWOPI                         023     153   0245   32 0048 0225
                NZE           CSV02                 024     154   0225   45 0228 0536
                BMI   CSV14                         025     155   0228   46 0245 0382
                FSB   ONEPI                         026     156   0382   33 0036 0263
                NZE   CSV16   CSV02                 027     157   0263   45 0066 0536
CSV15           FSB   TWOPI                         028     158   0096   33 0048 0275
                NZE           CSV02                 029     159   0275   45 0278 0536
                BMI           CSV15                 030     160   0278   46 0481 0096
                FAD   ONEPI                         031     161   0481   32 0036 0313
                NZE   CSV16   CSV02                 032     162   0313   45 0066 0536
CSV16           STU   CSV09                         033     163   0066   21 0020 0073
                RSU    8003                         034     164   0073   61 8003 0531
                STU   CSV10                         035     165   0531   21 0750 0503
                STU   CSV11                         036     166   0503   21 0108 0111
                LOO   FPONE                         037     167   0111   69 0040 0193
                STO   CSV12   CSV13                 038     168   0193   24 0065 0018
CSV13           RAU   CSV12                         039     169   0018   60 0065 0019
                FAD   FPONE                         040     170   0019   32 0040 0267
                STU   CSV17                         041     171   0267   21 0172 0325
                FAD   FPONE                         042     172   0325   32 0040 0317
                STU   CSV12                         043     173   0317   21 0065 0068
                RSU   CSV10                         044     174   0068   61 075U 0305
                FMP   CSV09                         045     175   0305   39 0020 0070
                FMP   CSV09                         046     176   0070   39 0020 0120
                FOV   CSV17                         047     177   0120   34 0172 0222
                FOV   CSV12                         048     178   0222   34 0065 0115
                STU   CSV10                         049     179   0115   21 0750 0553
                FOV   CSV11                         050     180   0553   34 0108 0158
                RAM    8003                         051     181   0150   67 8003 0165
                RAU    8002                         052     182   0165   60 8002 0123
                FSB   SIZE8                         053     183   0123   33 0076 0603
                BMI   CSV18                         054     184   0603   46 0056 0007
                RAU   CSV11                         055     185   0007   60 0108 0363
                FAD   CSV10                         056     186   0363   32 0750 0328
                STU   CSV11   CSV13                 057     187   0328   21 0108 0018
CSV18           RAU   CSV11   CSV02                 058     188   0056   60 0108 0536
FPONE            10    0000    0051                 059     189   0040   10 0000 0051
FPTWO            20    0000    0051                 060     190   0136   20 0000 0051
TWOPI            62    8318    5351                 061     191   0048   62 8318 5351
SIZE8            10    0000    0043                 062     192   0076   10 0000 0043
ONEPI            31    4159    2751                 063     193   0036   31 4159 2751
EOOLR           STO   EXP02                         001     194   0015   24 011U 0071
                STU   EXP03                         002     195   0071   21 0126 0079
                LOO    8007                         003     196   0079   69 8007 0185
                STO   EXP04                         004     197   0185   24 0038 0041
                RAC    0000                         005     198   0041   88 0000 0047
                RAU   EXP03                         006     199   0047   60 0126 0581
                BMI   EXP05   EXP06                 007     200   0581   46 0284 0235
EXP06           FSB   LNTEN                         008     201   0235   33 0088 0215
                BMI   EXP07                         009     202   0215   46 0168 0069
                AXC    0001   EXP06                 010     203   0069   58 0001 0235
EXP07           FAD   LNTEN   EXP08                 011     204   0168   32 0088 0255
EXP05           SXC    0001                         012     205   0284   59 0001 0240
                FAD   LNTEN                         013     206   0240   32 000U 0315
                BMI   EXP05   EXP08                 014     207   0315   46 0284 0265
EXP08           STU   EXP03                         015     208   0265   21 0186 0129
                STU   EXP09                         016     209   0129   21 0334 0437
                RAL   EXP04                         017     210   0437   65 0038 0843
                LOO    8007                         018     211   0243   69 8007 0049
                STO   EXP04                         019     212   0049   24 0038 0091
                RAC    8002                         020     213   0091   88 8002 0099
                LOD   FPONE                         021     214   0099   69 0040 0893
                STO   EXP10                         022     215   0293   24 0146 0149
                STD   EXP11   EXP12                 023     216   0149   24 0108 0355
EXP12           RAU   EXP10                         024     217   0355   60 0146 0151
                FAD   EXP09                         025     218   0151   32 0334 0161
                STU   EXP10                         026     219   0161   21 0146 0199
                RAU   EXP11                         027     220   0199   60 0102 0057
                FAD   FPONE                         028     221   0057   32 0040 0367
                STU   EXP11                         029     222   0367   21 0102 0405
                RAU   EXP09                         030     223   0405   60 0334 0239
                FMP   EXP03                         031     224   0239   39 0126 0176
                FOV   EXP11                         032     225   0176   34 0102 0152
                STU   EXP09                         033     226   0152   21 0334 0487
                FOV   EXP10                         034     227   0487   34 0146 0196
                FSB   SIZE8                         035     228   0196   33 0076 0653
                BMI           EXP12                 036     229   0653   46 0106 0355
                RAU   EXP10                         037     230   0106   60 0146 0201
                AUP   EXP04   EXP02                 038     231   0201   10 0038 0118
FPONE            10    0000    0051                 039     232   0040   10 0000 0051
SIZE8            10    0000    0043                 040     233   0076   10 0000 0043
LNTEN            23    0258    5151                 041     234   0088   23 0258 5151
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RESULTS AND DISCUSSION

Steady-state Tubular Flow Reactor

The concentration profiles for the various combinations of M and R for the first-order chemical reactions were calculated by use of equations (9) through (12), (16) and (18). Typical results corresponding to R=1/2 for B.C.(I), (II) and (III) are shown in Fig. 1 and those corresponding to R=2 for B.C.(IV) are shown in Fig. 2. The figures show that the concentration profiles obtained by use of B.C.(II), (III) and (IV) were inconsistent with the limits of both a plug flow reactor and a uniformly stirred tank reactor for all values of M and R; whereas the profiles computed by use of B.C.(I) were consistent with such limits.

The word "inconsistent" means that a computed conversion or concentration profile either less than that of a uniformly stirred tank reactor or greater than that of a plug flow reactor is unreasonable. Models which predict this are therefore "inconsistent". The numerical values of the limits of inconsistency were obviously calculated by use of equations (16) and (18) for a uniformly stirred tank reactor and a plug flow reactor respectively.

Figure 3, however, indicates that the over-all conversions computed by use of the different sets of boundary conditions were numerically in agreement for large values of M (small degree of axial dispersion) and deviate increasingly from each other as the degree of axial dispersion increases. Again the overall conversions computed by use of B.C.(II) and (III) gave values lower than those obtained for a uniformly stirred tank reactor, and those

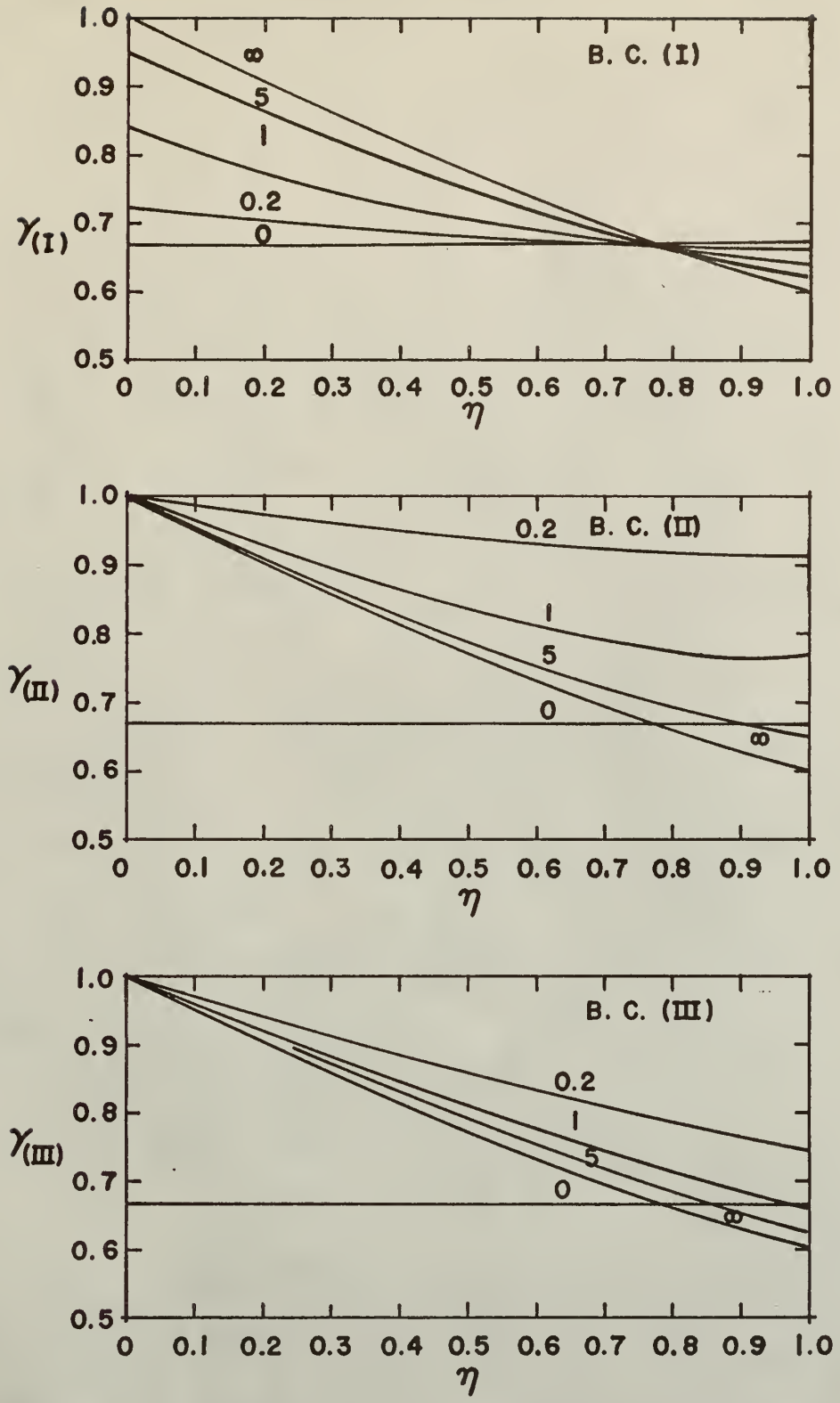

FIG. 1. Concentration profile of reactant for various M for first-order reaction with $R = \frac{1}{2}$.

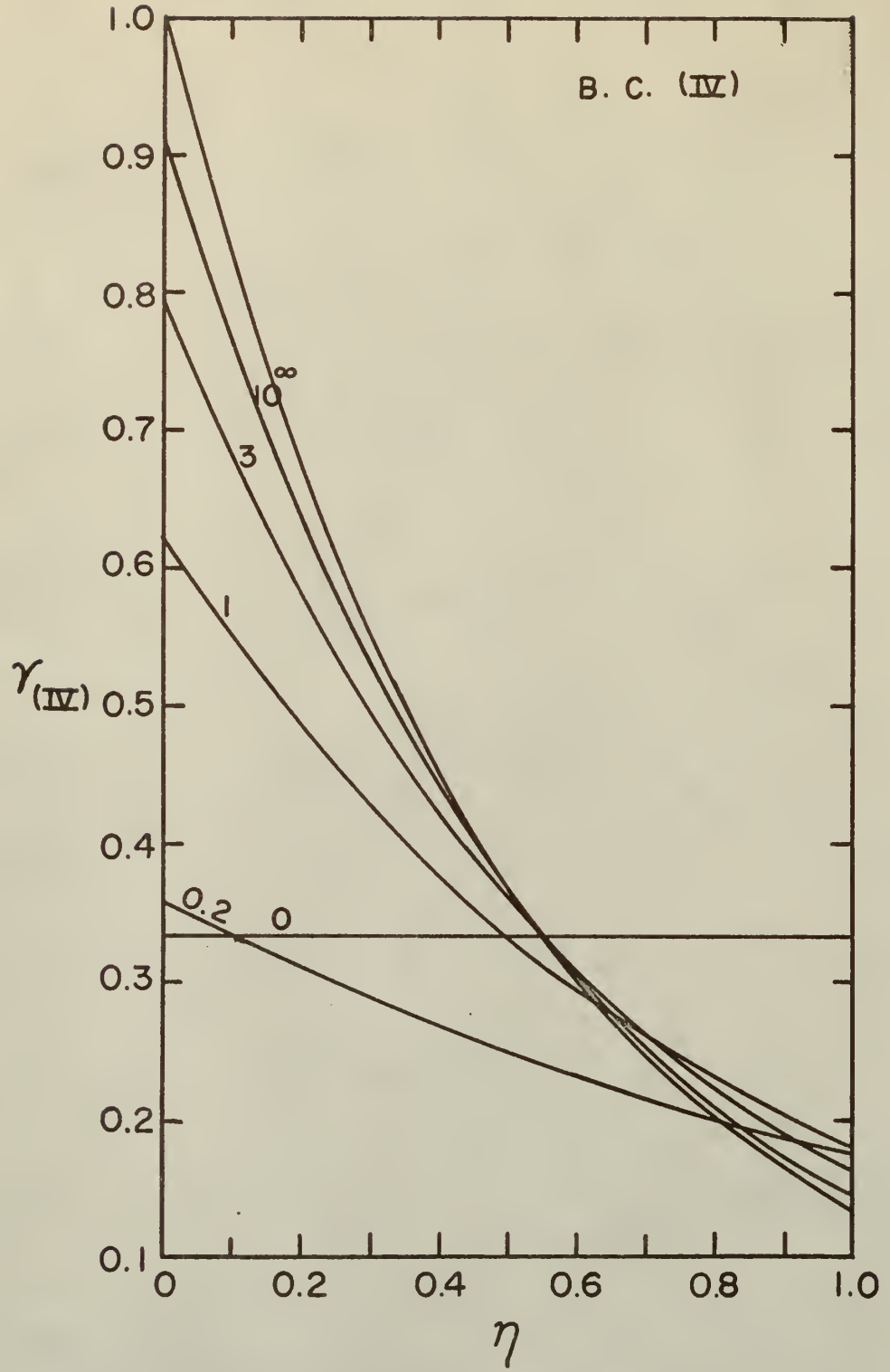

Fig. 2. Concentration profile of reactant for various M for first-order reaction with R=2

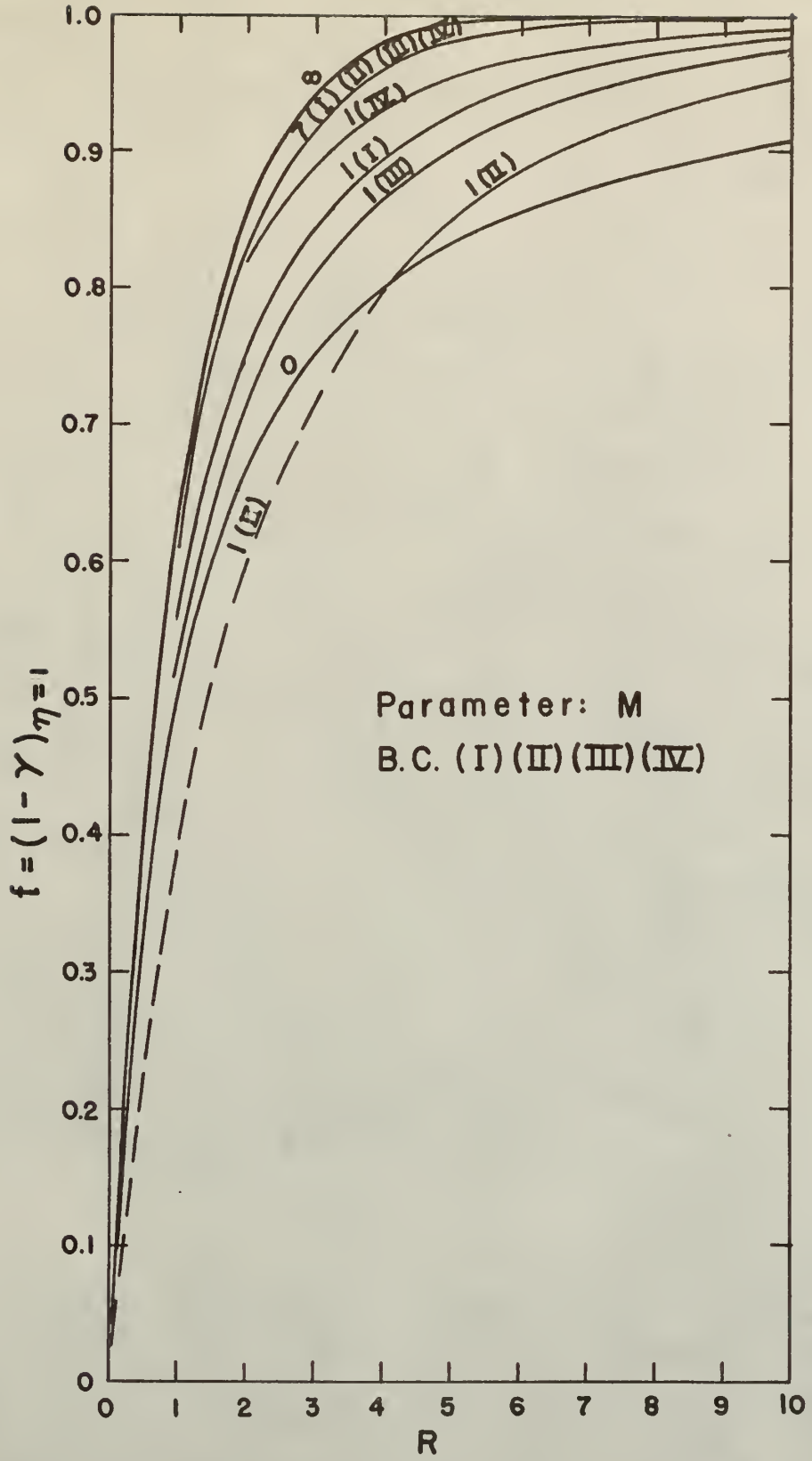

FIG. 3. Comparison of final conversions as a function of R for first-order reaction.

computed by use of B.C.(IV) gave values higher than those obtained in a plug flow reactor, especially for low conversions and a high degree of axial dispersion, i. e., low values of R and M. Reactors falling in this category are sometimes met in laboratory and pilot plant experimentation, though not commonly required for design purposes.

The inconsistency shown in Figs. 1 and 2 led to the establishment of the domain for which any combinations of M and R are inconsistent, with the limits of the domain, as shown in Figs. 4 and 5. The curves in Fig. 4 show the locus representing:

$$(f_{II})_{\eta=1} - f_{M=0} = 0$$

$$(f_{III})_{\eta=1} - f_{M=0} = 0$$

The domain below the curves, then, corresponds to the condition:

$$(f_{II})_{\eta=1} - f_{M=0} < 0$$

$$(f_{III})_{\eta=1} - f_{M=0} < 0$$

The curve in Fig. 5 indicates the locus representing:

$$(f_{IV})_{\eta=1} - f_{M=\infty} = 0$$

The domain below the curve, then, corresponds to the condition:

$$(f_{IV})_{\eta=1} - f_{M=\infty} > 0$$

Where f represents the overall conversions.

The inconsistency of the overall conversions using B.C.(II) has been indicated by Wehner and Wilhelm (24). However, it has

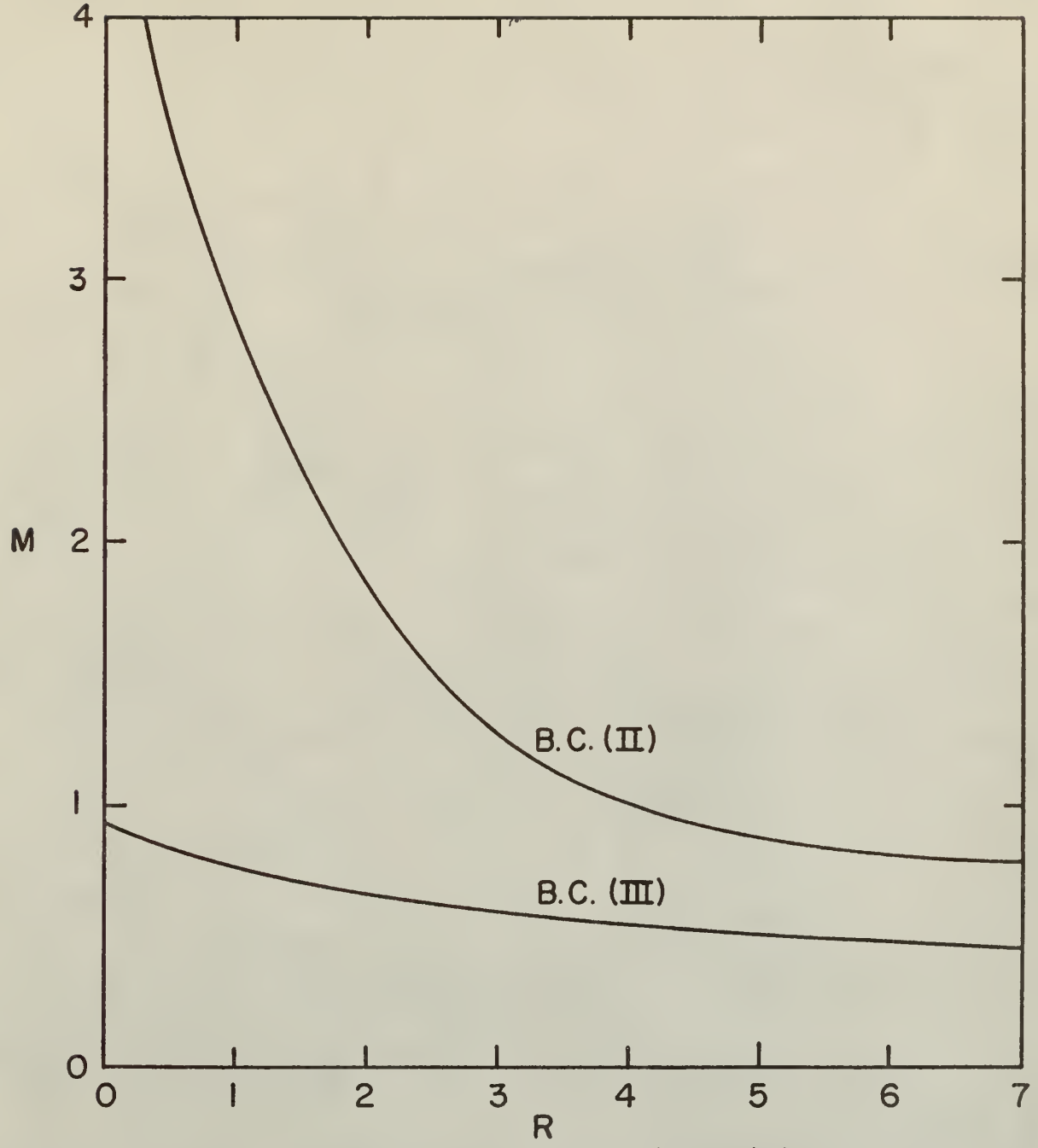

Fig. 4. Locus indicating regions in which conversions are inconsistent with limit of an uniformly stirred tank reactor for first-order reaction.

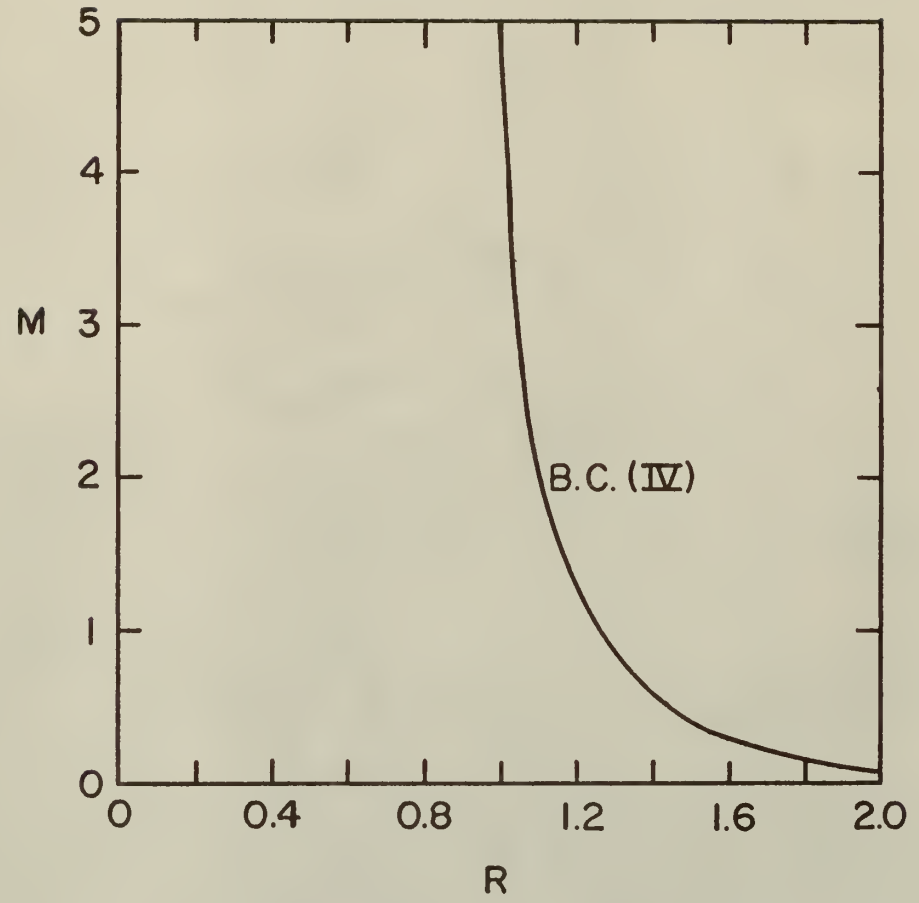

Fig. 5. Locus indicating regions in which conversions are inconsistent with limit of a plug flow reactor for first-order reaction.

never been established quantitatively as a function of the system characteristics, i. e., M and R. The cases of B.C.(III) and (IV) have been hitherto unknown. The overall conversions computed by use of B.C.(II) and (III) are consistent with the upper limit of a plug flow reactor, and those by use of B.C.(IV) are consistent with the lower limit of a stirred tank reactor.

The design of an isothermal reactor should not be based only on the overall conversion when the reactions to be carried out in the reactor are exothermic or endorthermic. The estimation of the heat transfer area to be provided requires a knowledge of the concentration profile, or the conversion along the length of reactor. Figs. 6 and 7 show the comparison of concentration profiles based on the different sets of boundary conditions at both high and low degree of dispersions. The figures supplement the previous statement that the deviations among the profiles decrease as M increases. This means that the effect of boundary conditions become negligible for high values of M, say of the order of 15, and the reactor can be assumed to behave as a plug flow reactor with reasonable accuracy. The percentage deviations of the overall conversions computed by use of B.C.(II), (III) and (IV) from those computed by use of B.C.(I) are shown in Figs. 8, 9 and 10. The reason that B.C.(I) is used as a basis for the computation is that, as pointed out previously, this yields conversions which are consistent with both limits of the reactor. These figures show that the deviations increase as M and R decrease, and B.C.(II) always yields larger deviations than B.C.(III) and (IV). Since the solutions to B.C.(III) and (IV) are in the form of a simple exponential

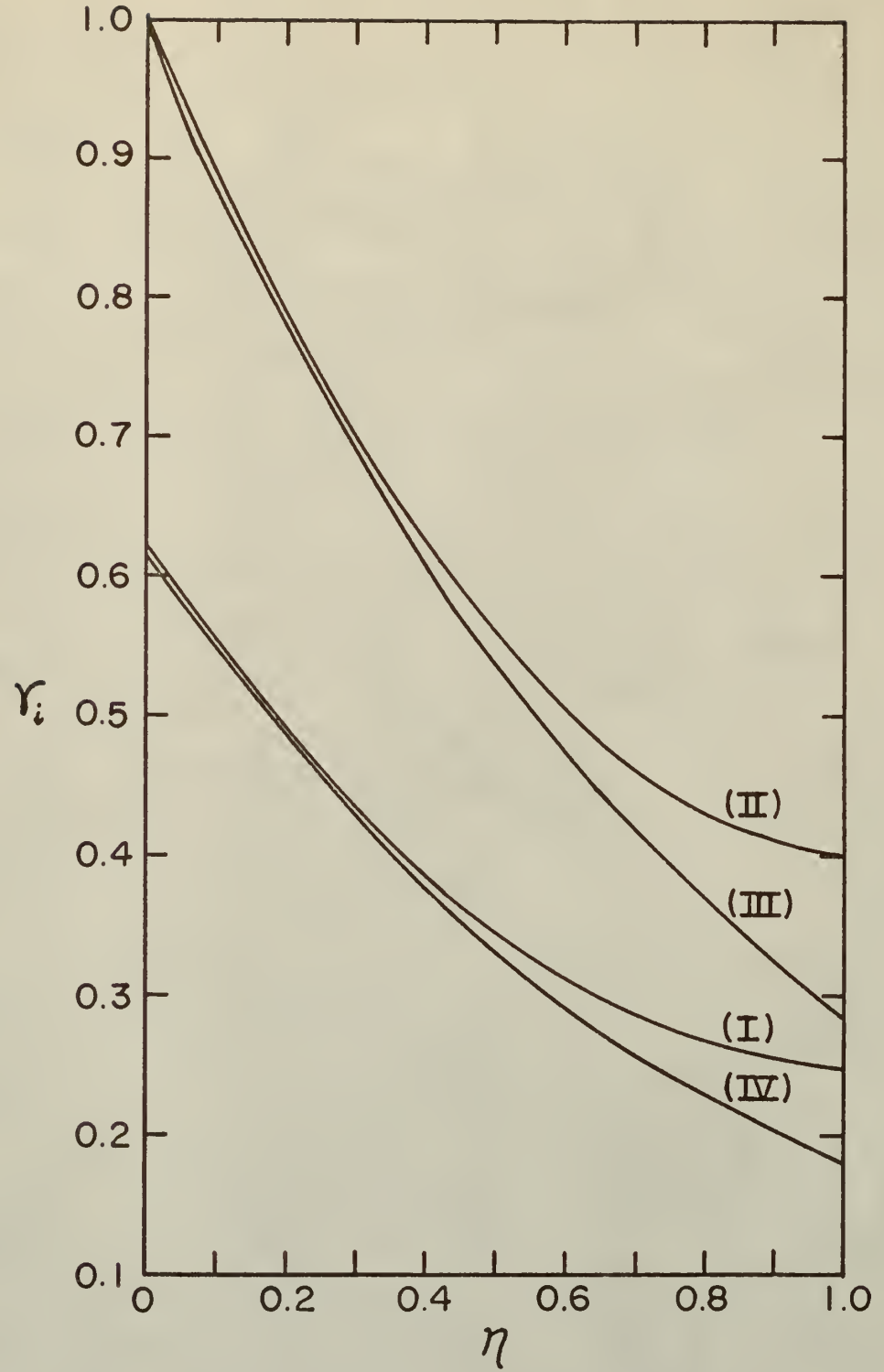

Fig. 6. Comparison of concentration profile of reacting systems (R=2) based on different boundary conditions at high degree of dispersion (M=1).

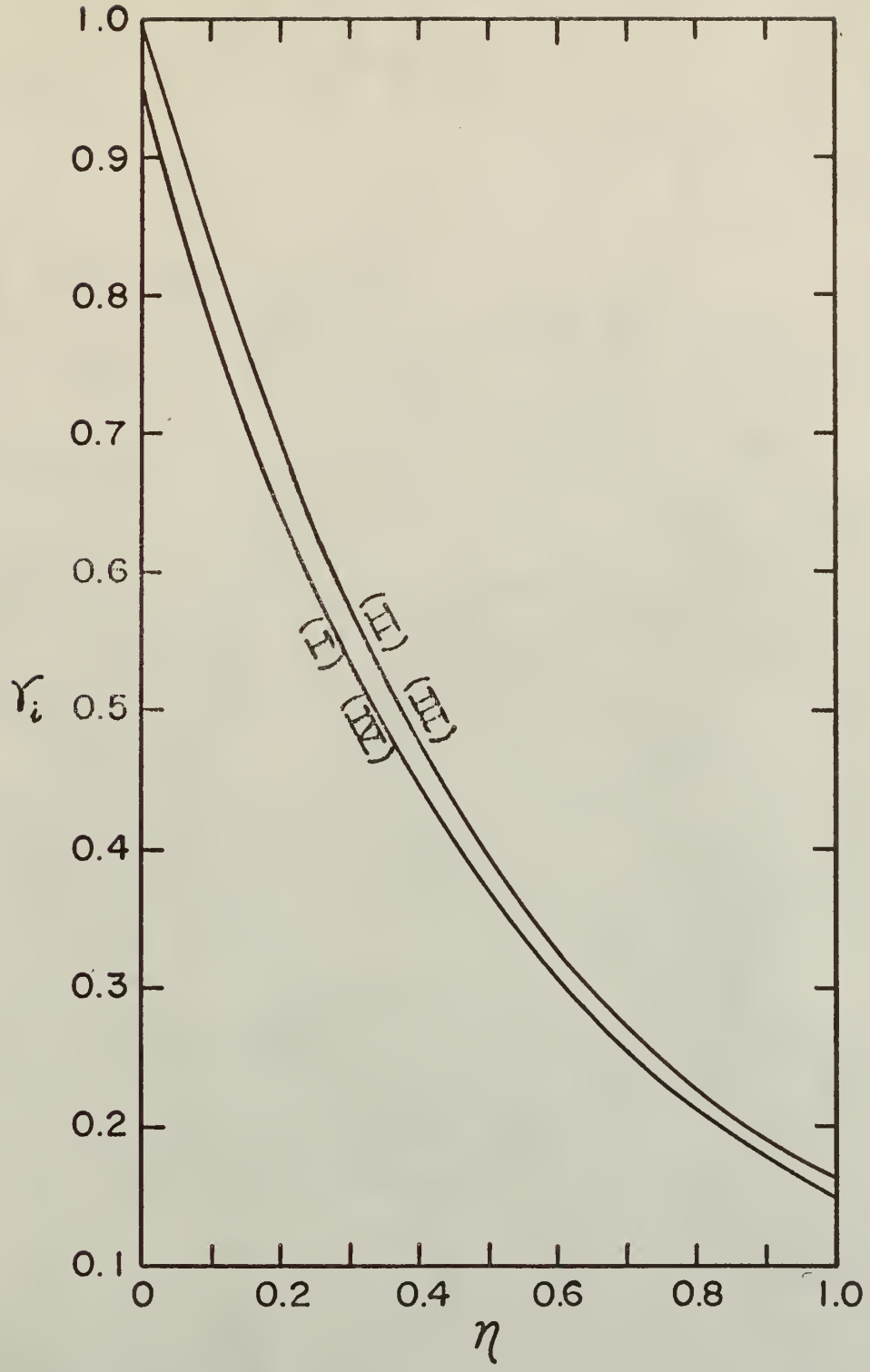

Fig. 7. Comparison of concentration profile of reacting systems (R=2) based on different boundary conditions at low degree of dispersion (M=15).

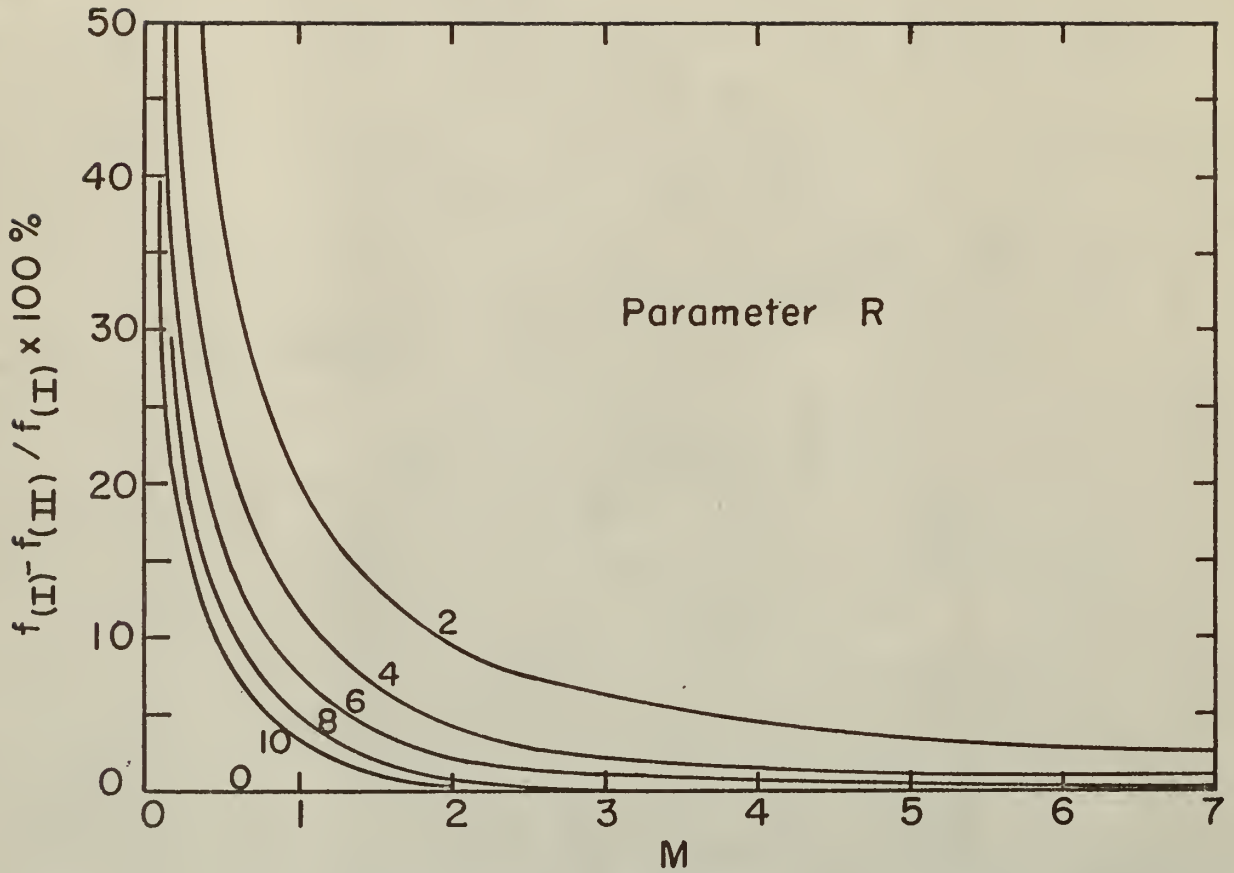

Fig. 8. Percentage deviations of final conversions computed by use of boundary conditions (II) from those computed by use of boundary conditions (I) as functions of M for first-order reactions.

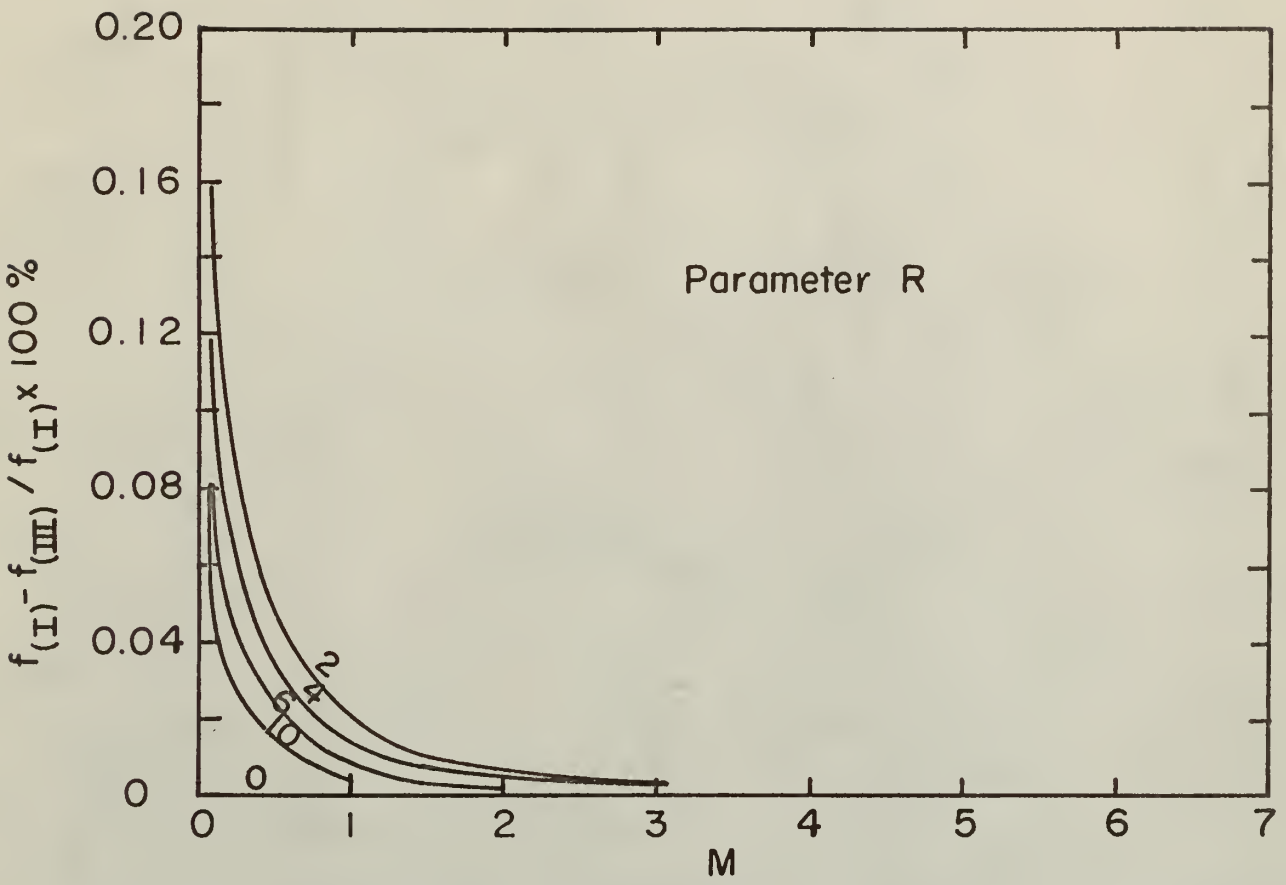

Fig. 9. Percentage deviations of final conversions computed by use of boundary conditions (III) from those computed by use of boundary conditions (I) as functions of M for first-order reactions.

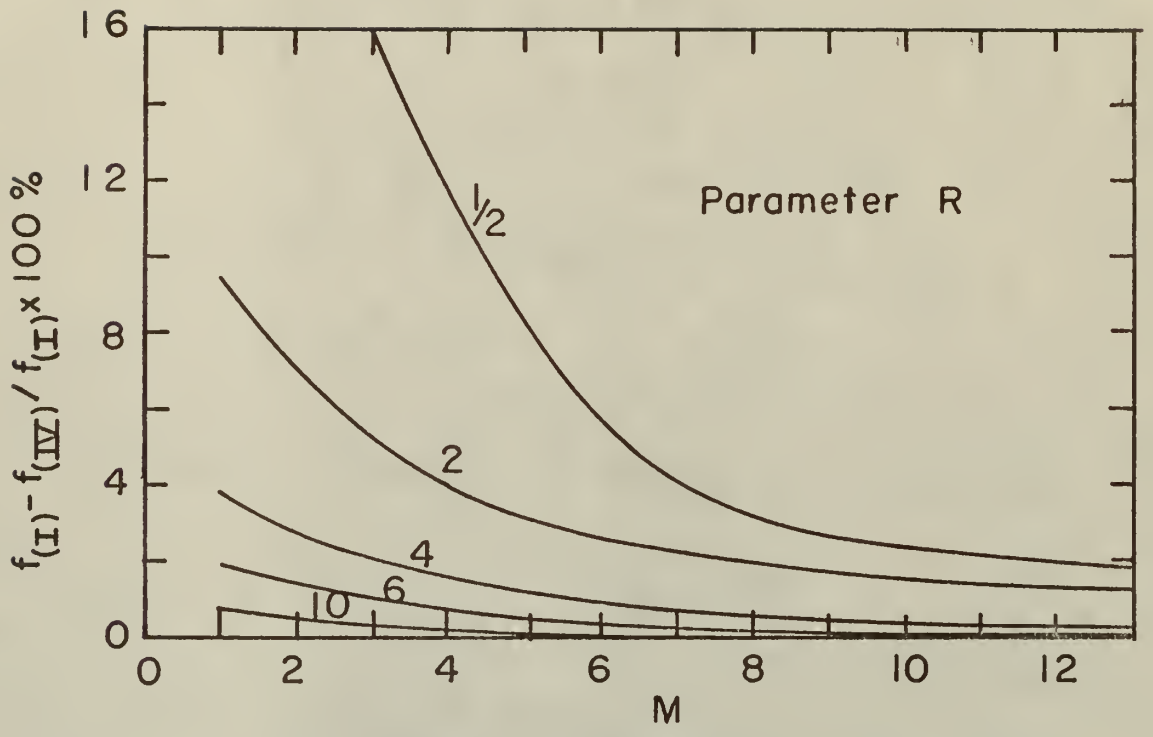

Fig. 10. Percentage deviations of final conversions computed by use of boundary conditions (IV) from those computed by use of boundary conditions (I) as functions of M for first-order reaction.

function, their use to approximate B.C.(I) is always more convenient, and also yields more accurate results than the use of B.C.(II).

The establishment of regions of inconsistency for the use of different sets of boundary conditions gives a set of criteria for the use of B.C. (II), (III) and (IV). These boundary conditions should be avoided in the domain of inconsistency for computing final conversions. Due to ease of solving simple exponential functions, the use of B.C.(III) or (IV), if their limits are recognized, offers the most convenient means for computing the final conversions. The recognition of the limit, of course, refers to the domain above the curves in Figs. 4 and 5. The use of B.C.(II) for a homogenous tubular flow reactor with axial dispersion is not desirable from the view point of both accuracy and convenience. Though the use of B.C.(IV) has never been attempted previously, it is useful in the computation of conversions around the inlet and center of the reactor, since as shown in Figs. 6 and 7, the concentration profile computed by use of B.C.(IV) follows closely that computed by the use of B.C.(I), except at the exit of reactor. As stated previously, the simple analytical solution in exponential form for B.C.(IV) provides a reasonably accurate and convenient method of evaluating conversions.

Unsteady-state Tubular Flow Reactor

The numerical calculations of the impulse response function tabulated in Table 4 are relatively easy in comparison with the direct numerical solution of the original partial differential equation, equation (2). Some computational results of the impulse

response functions, both with and without chemical reaction, are presented in Figs. 11 through 18. These figures definitely indicate the differences of response based on different sets of boundary conditions. Four sets of impulse response curves at R=0 for high and low degree of dispersion are replotted in Figs. 19 and 20 to show that as the degree of dispersion becomes small, say of the order of M=15, all the response curves representing the four boundary conditions approximately converge to one and the deviations among the four response curves increase as degree of dispersion increases. This fact supplements the previous statement that the effect of boundary conditions becomes negligible above the value of M=15. Exactly the same observations are given in Figs. 15 through 18 where the response curves of first-order chemical reaction are considered, except that the magnitude of the response curves decrease rapidly with increase in R. This is due to the fact that the magnitude of R is proportional to the rate of reaction; thus the faster the reaction the faster the concentration decreases.

One application of such computed response curves is the determination of the axial dispersion coefficient from a dynamic experiment. A survey of published literature shows that most of the methods used to determine the mean dispersion coefficient from transient response experiments makes use of only a single point of an entire experimental response curve, such as the maximum point or the slope at unit time (4) (25). It is known that the use of a single datum in drawing conclusions for an entire experiment may lead to serious error because of probable experimental error.

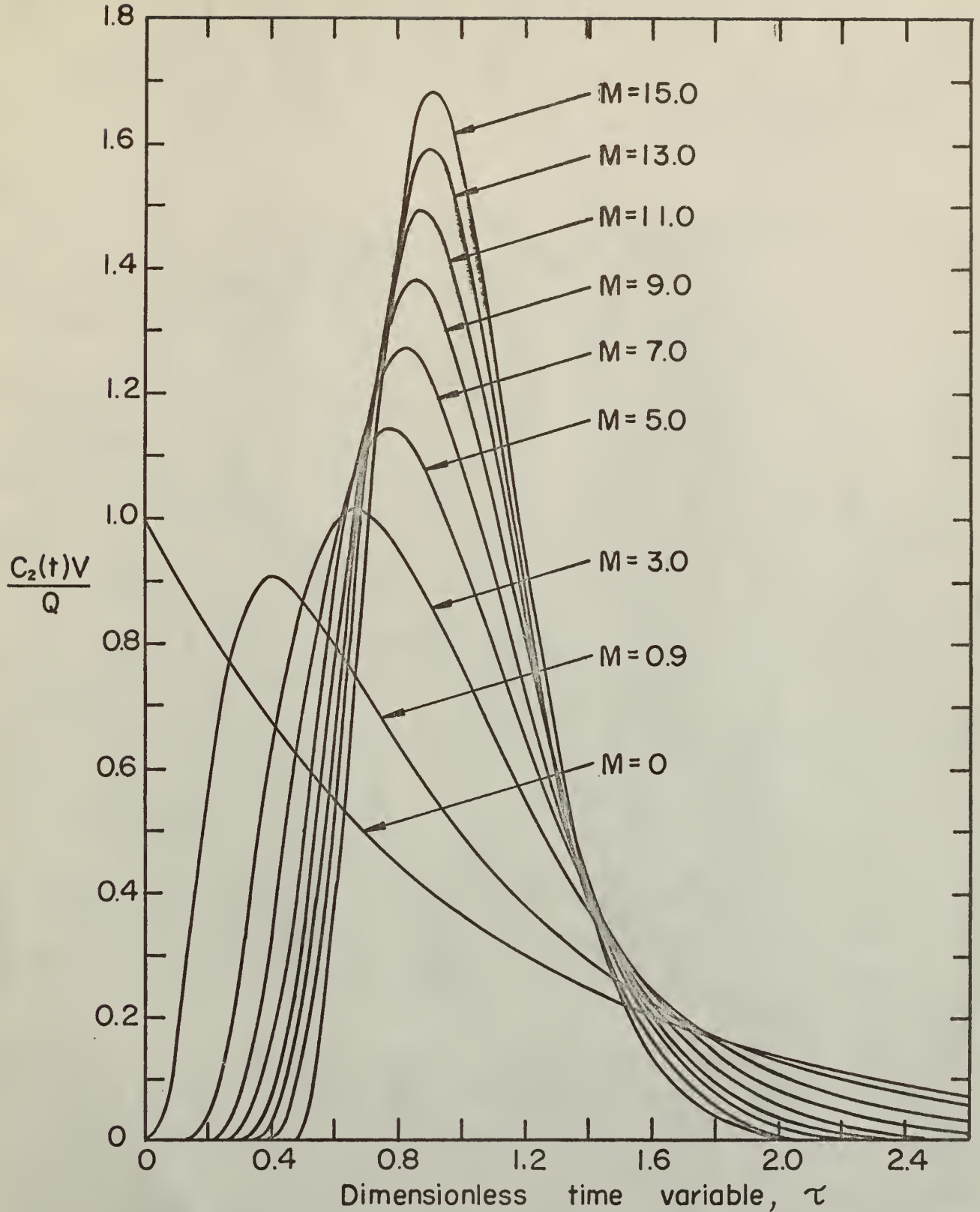

Fig. 11. Calculated response curves of exit concentration against impulse for non-reacting systems (R=0) based on B. C. (I) with M as parameter.

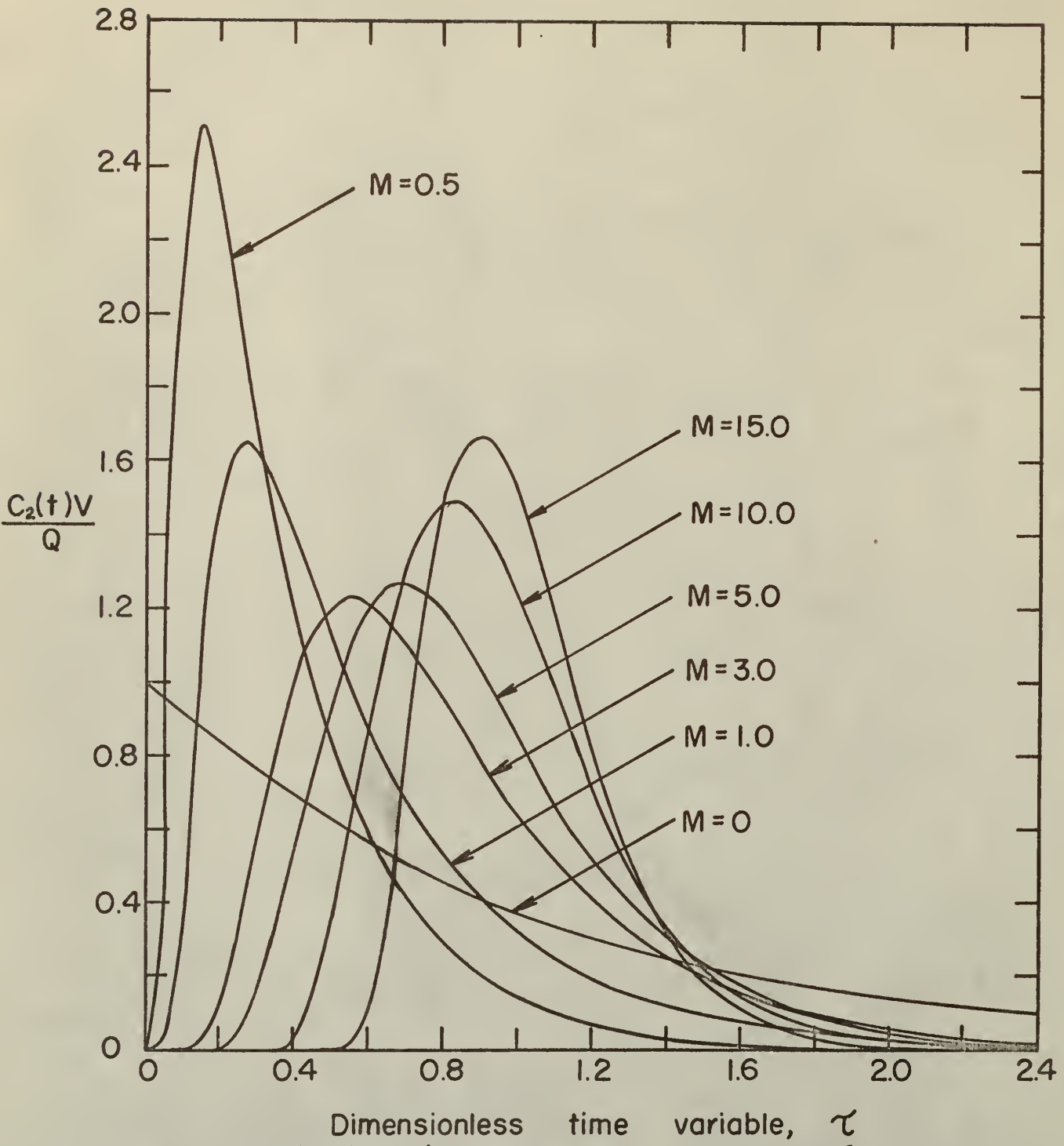

Fig. 12. Calculated response curves of exit concentration against impulse for non reacting systems (R=0) based on B. C. (II) with M as parameter.

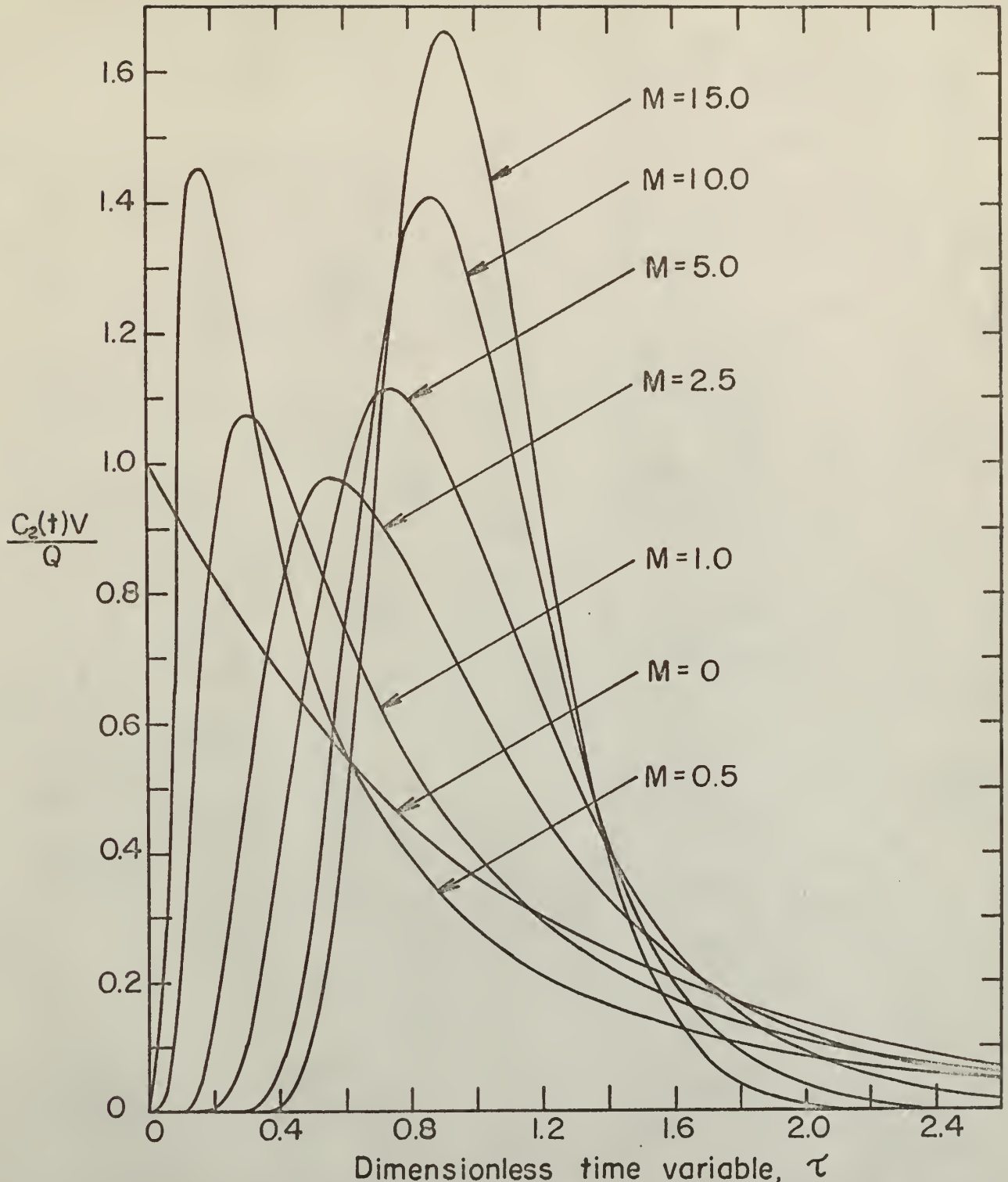

Fig. 13. Calculated response curves of exit concentration against impulse for non-reacting systems (R=0) based on B. C. (III) with M as parameter.

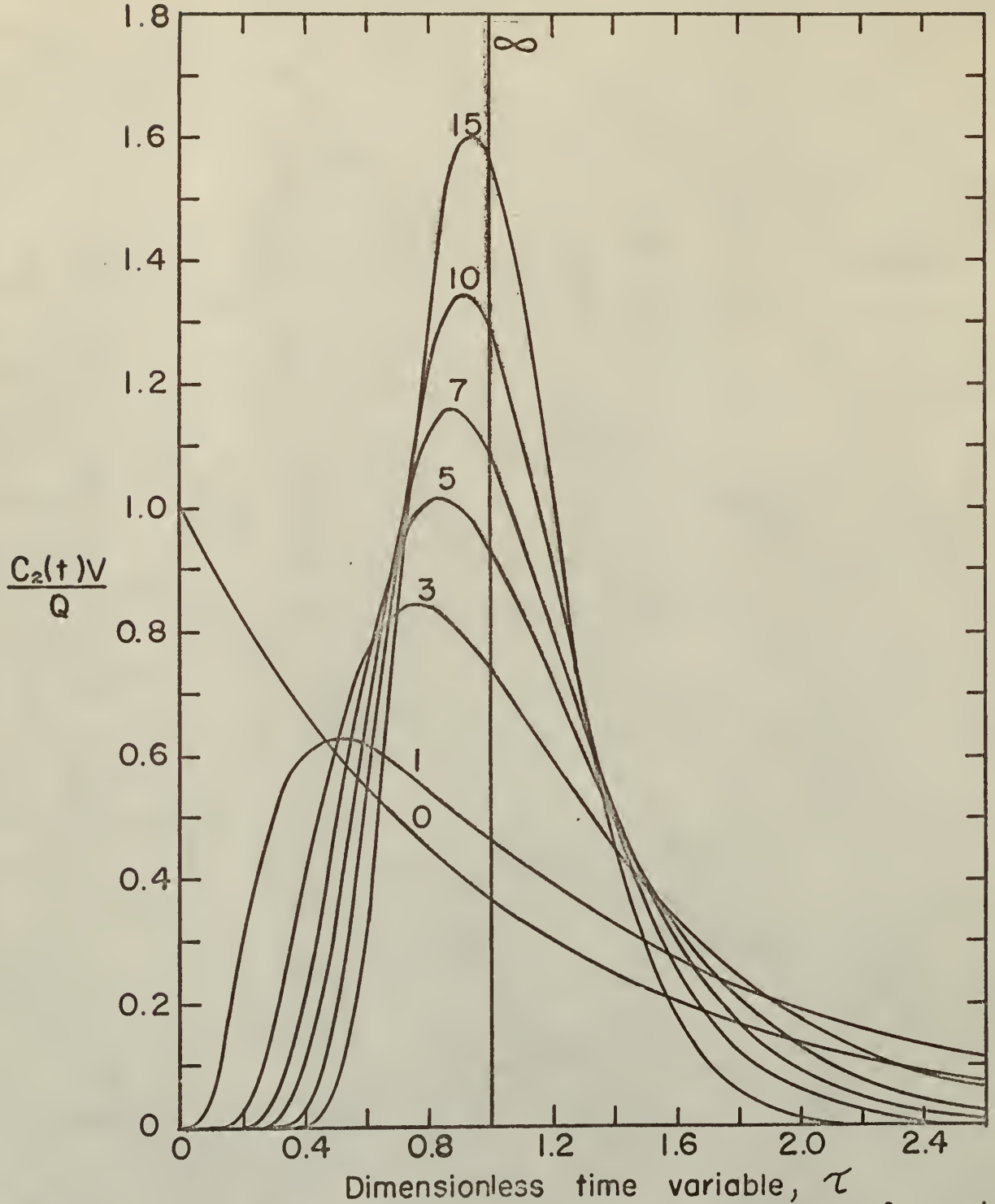

Fig. 14. Calculated response curves of exit concentration against impulse for non-reacting systems (R=0) based on B. C. (IV) with M as parameter.

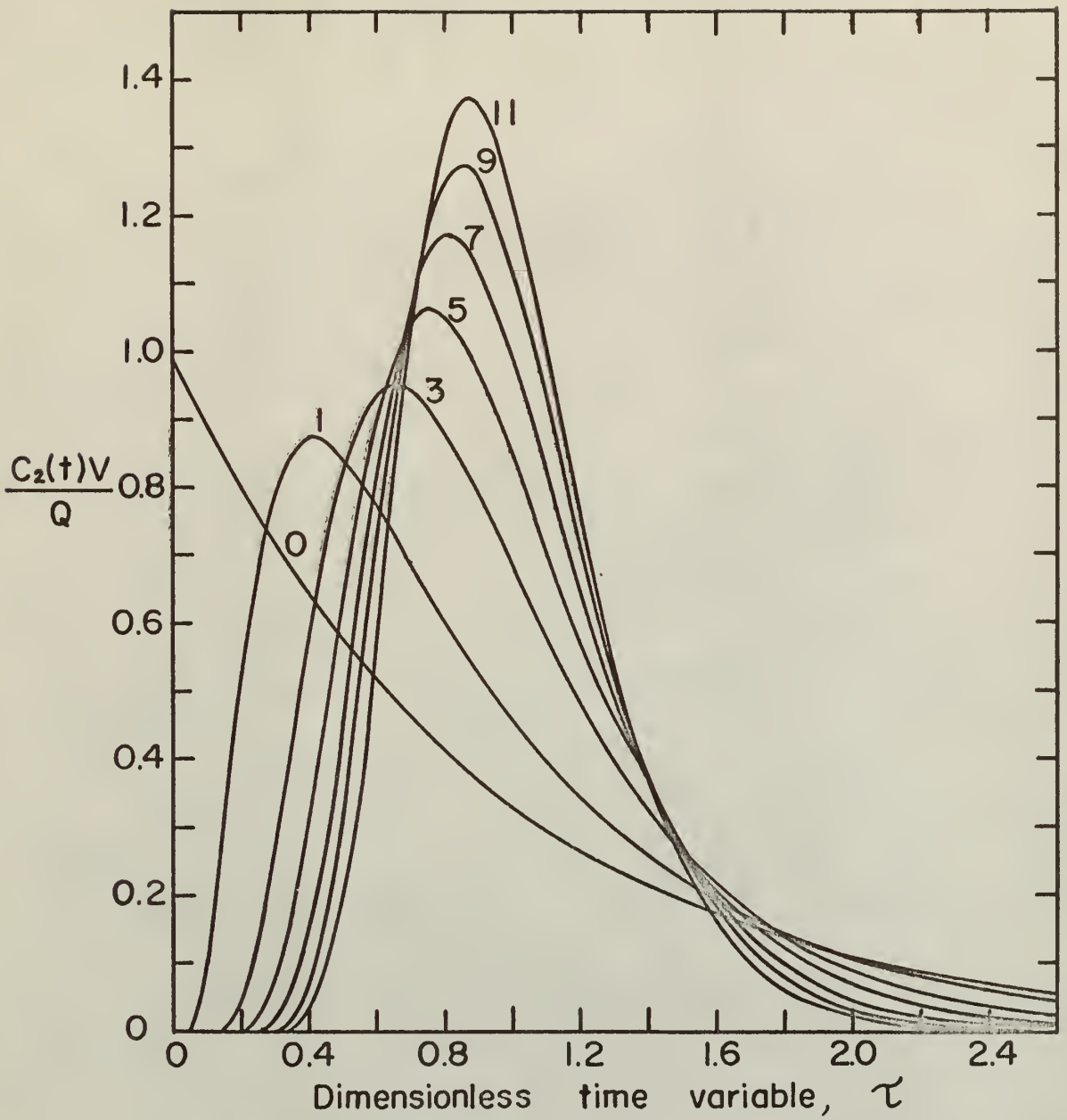

Fig. 15. Calculated response curves of exit concentration against impulse for reacting systems (R=0.1) based on B. C. (I) with M as parameter.

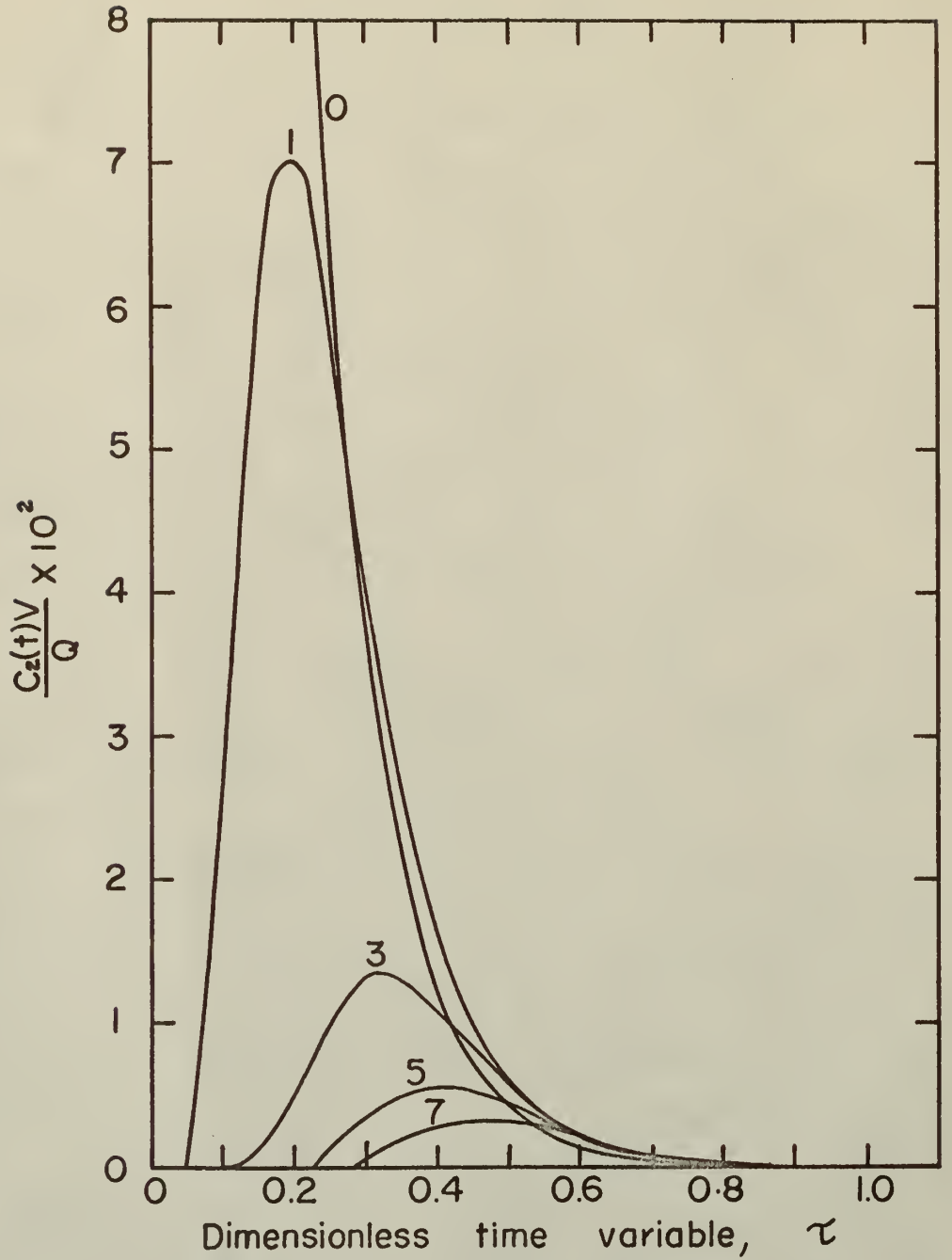

Fig. 16. Calculated response curves of exit concentration against impulse for reacting systems (R=10) based on B.C. (I) with M as parameter.

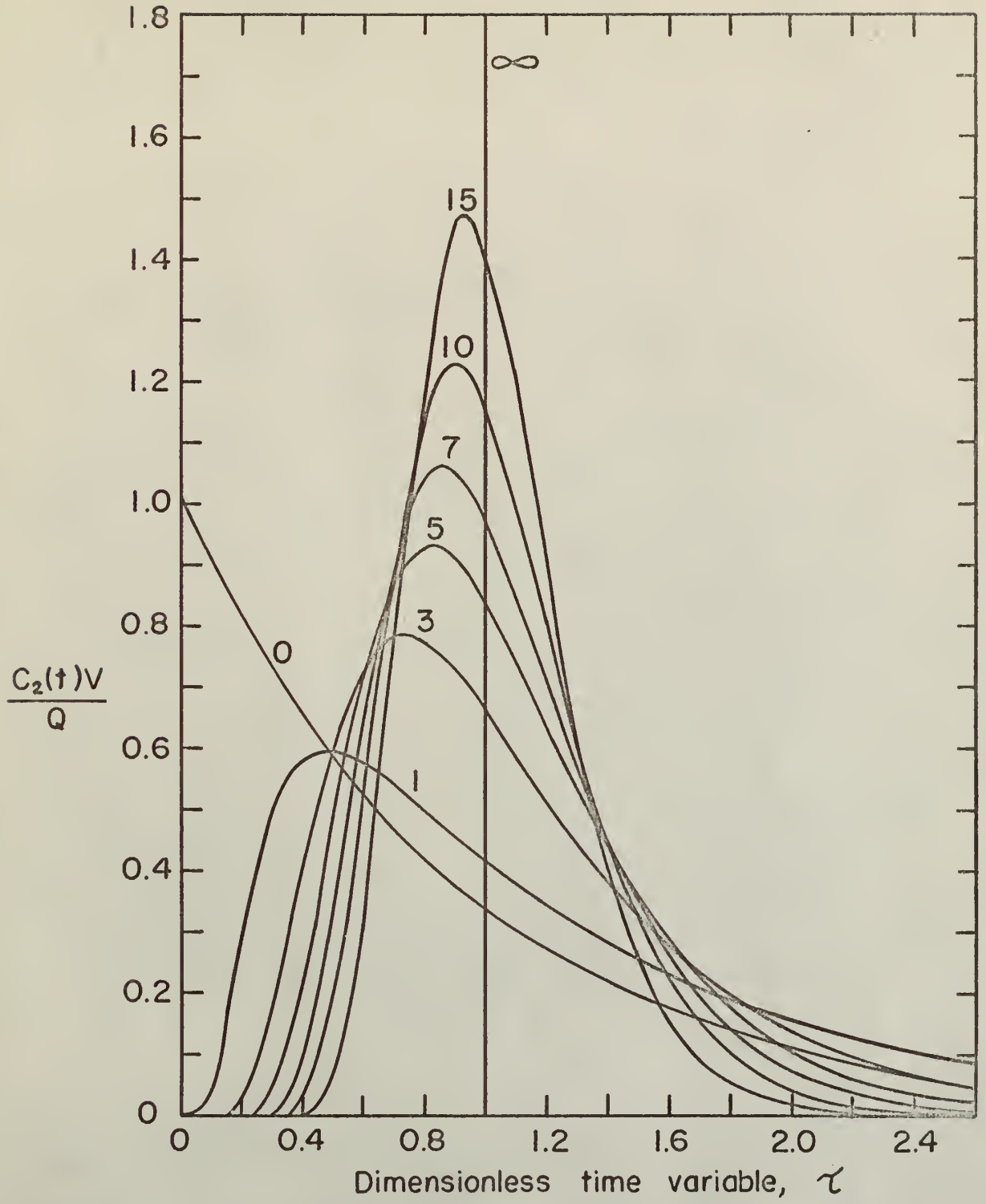

Fig. 17. Calculated response curves of exit concentration against impulse for reacting systems (R=0.1) based on B.C. (IV) with M as parameter.

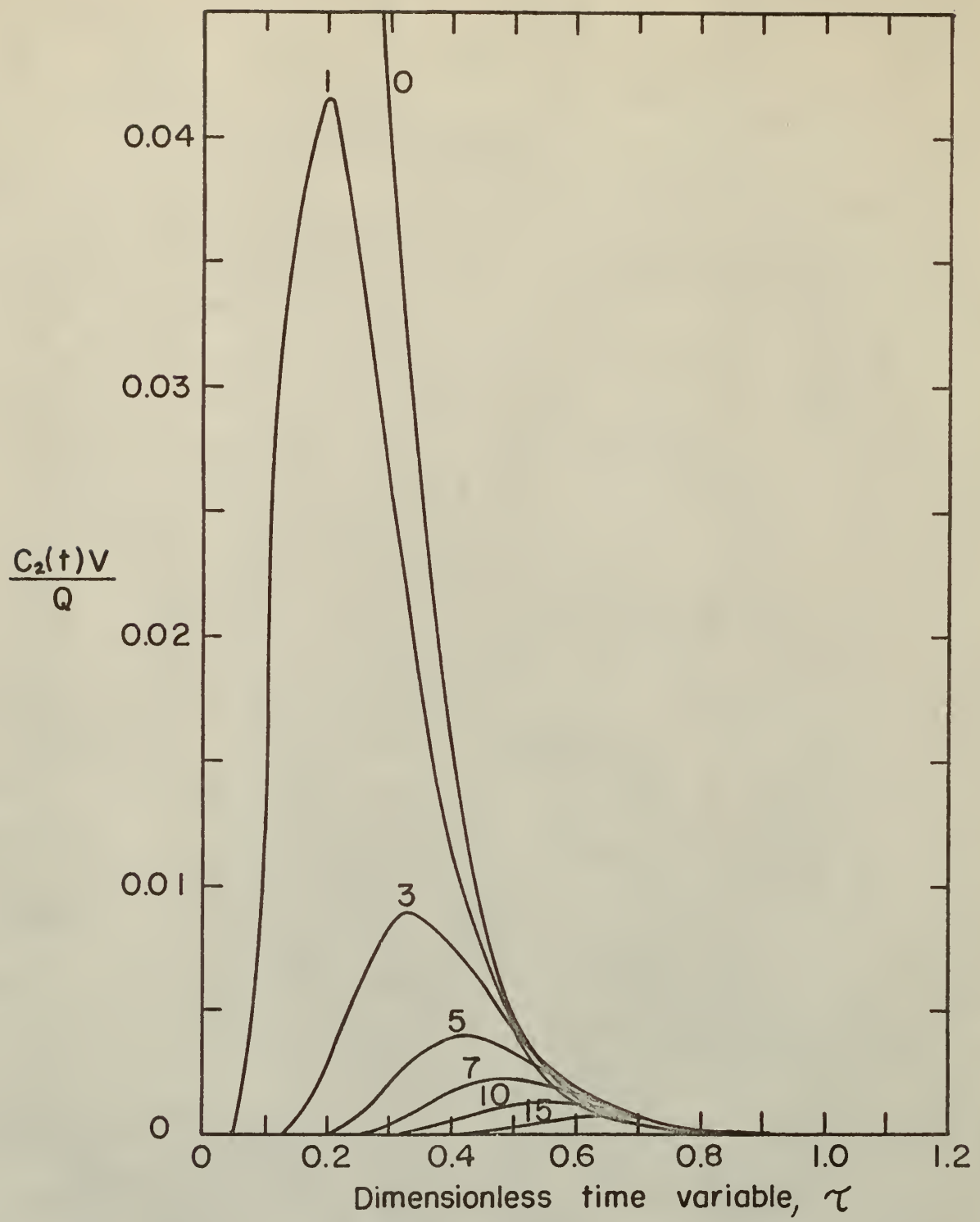

Fig. 18. Calculated response curves of exit concentration against impulse for reacting systems (R=10) based on B.C. (IV) with M as parameter.

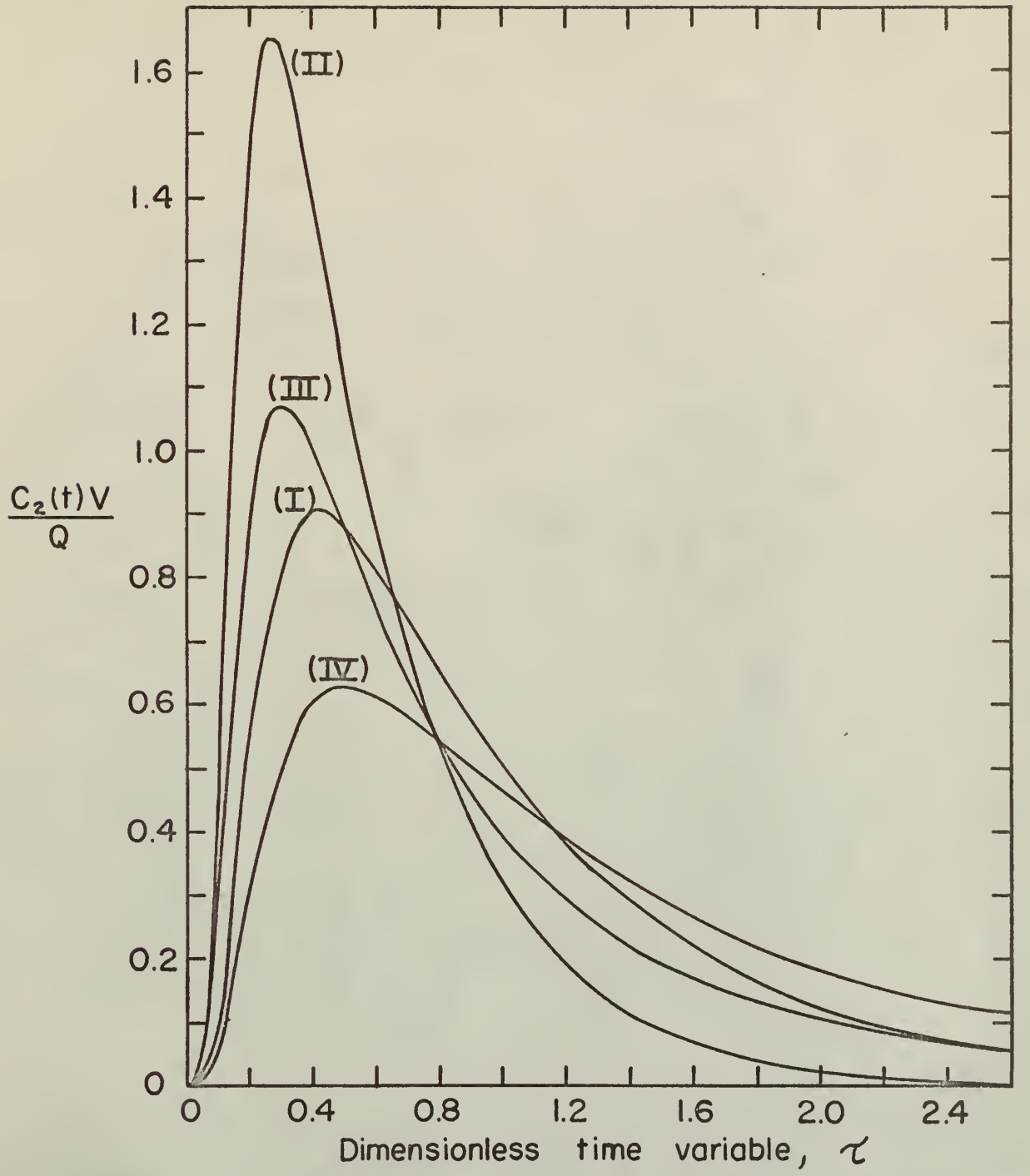

Fig. 19. Comparisons of response to impulse for non reacting systems (R=0) based on different boundary conditions at high degree of dispersion (M=1).

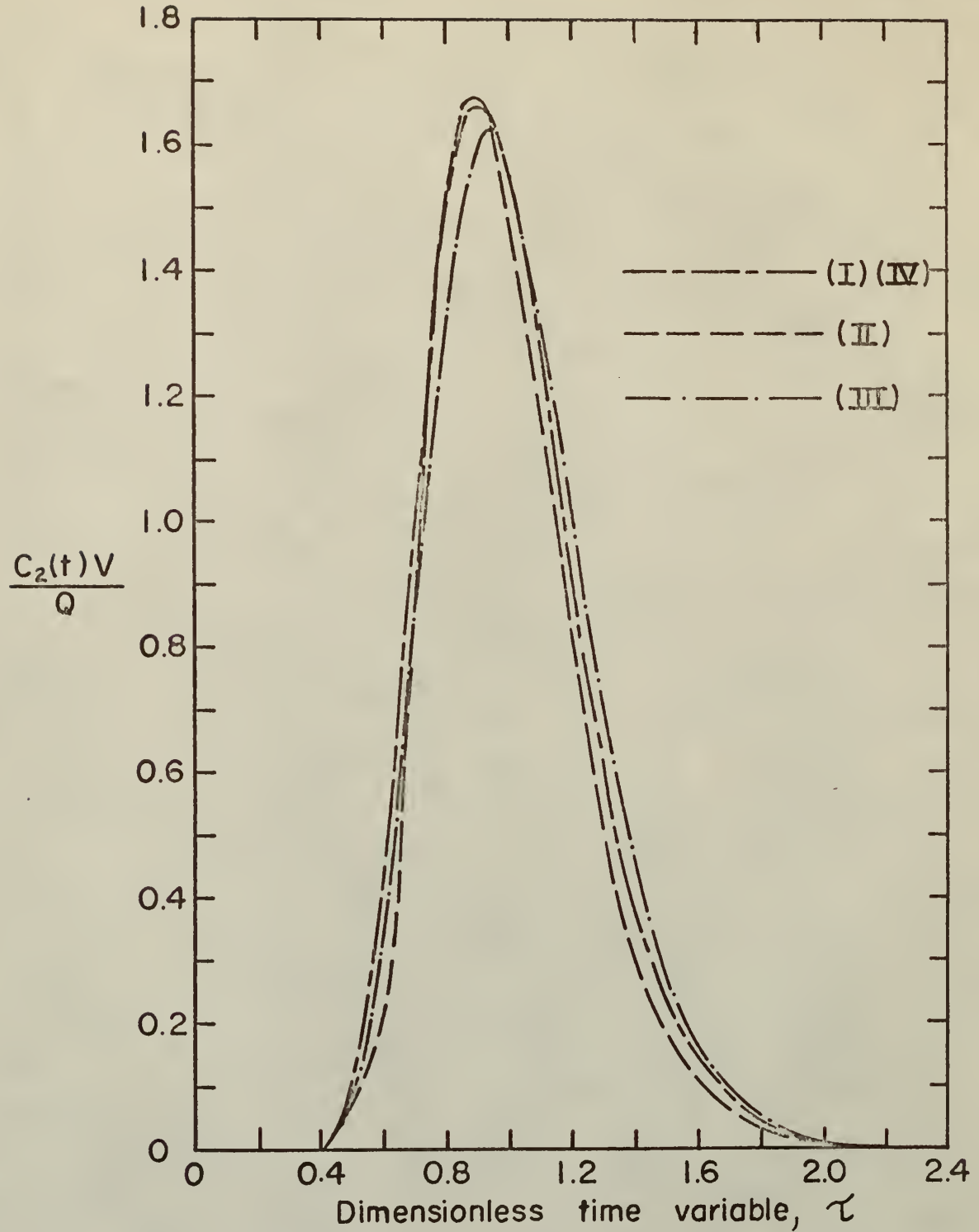

Fig. 20. Comparisons of response to impulse for non-reacting systems (R=0) based on different boundary conditions at low degree of dispersion (M=15).

Another approach in estimating $\overline{D}$ is reported by van der Laan (21), Levenspiel and Smith (12) who introduced the concept of the variance of the concentration distribution by momentum analysis. This method has the advantage by avoiding the cumbersome back transformation of the Laplace transform. The disadvantage, however, is that one cannot explain how distribution functions for residence times can be defind and measured for actual systems. Since the overall flow behavior problem is often of prime concern in the design of continuous processing equipment, one should compare response curves calculated from the assumed models and boundary conditions with the experimental curves.

Since any model of fluid dispersions, including the diffusion-type model, is approximate in nature, the best procedure for estimating the mean dispersion coefficient based on any one of the models should be the comparison of the calculated response curves and an experimental response curve. Such a procedure is illustrated in Fig. 21.

The experimental data used in Fig. 21 were obtained from Run 194 of Fowler and Brown (6). Levenspiel and Smith (12) obtained the value of mean dispersion coefficient of 2.28 $ft^2/sec.$ for this experiment by the methods of both maximum point and variance of concentration distribution curve. Inspection of Fig. 21, however, shows that the comparison of the entire response curves yields mean dispersion coefficient of 2.7 $ft^2/sec.$ if the response curve based on B.C.(III) is used. It also indicates that the system represented by the experimental data may not be represented by B.C.(II) and (IV). In other words, the comparison of the

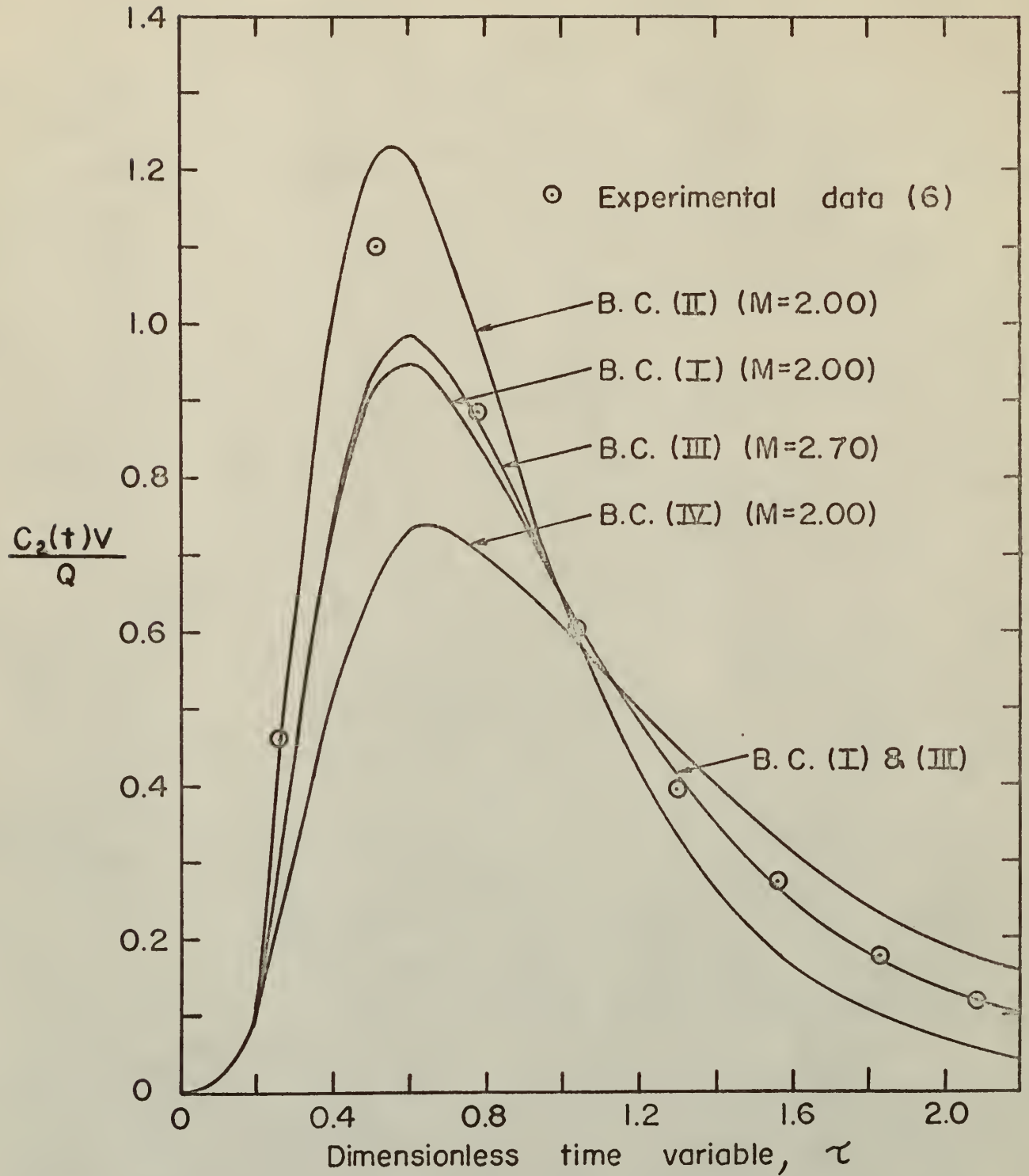

Fig. 21. Comparison of experimental and calculated response curves.

experimental and calculated response curves should lead to the establishment of the appropriate models or boundary conditions.

There are no data available on impulse experiments accompanied by first-order chemical reaction. If the data were available, the calculated response curves based on first-order chemical reaction (Figs. 15 through 18) could be useful to extract transport parameters from a dynamic experiment.

The typical results for step input response curves are shown in Figs. 22 through 25 both with and without chemical reaction. Due to the consistency observed in the steady-state concentration profile with the limits of both stirred tank and plug flow reactor, only the response curves based on B.C.(I) are presented here. The response curves with first-order chemical reaction do not approach one as τ approaches infinity because of the steady-state conversion, i.e., the response curves approach steady-state conversion as τ approaches infinity.

One method of determining $\bar{D}$ from step input response curves is illustrated by Levenspiel and Smith (12). They replotted the data of Von Rosenberg (23) on probability paper to obtain a straight line. From the property of normal distribution that the interval between the 16th and 84th percentile points is two standard deviations, they evaluated the mean dispersion coefficient. This method requires an extra step, and also involves appreciable error because the actual experimental data does not give a symmetric normal distribution curve. The best procedure for determining $\bar{D}$, again therefore, should be by matching the calculated response curves and an experimental response curve.

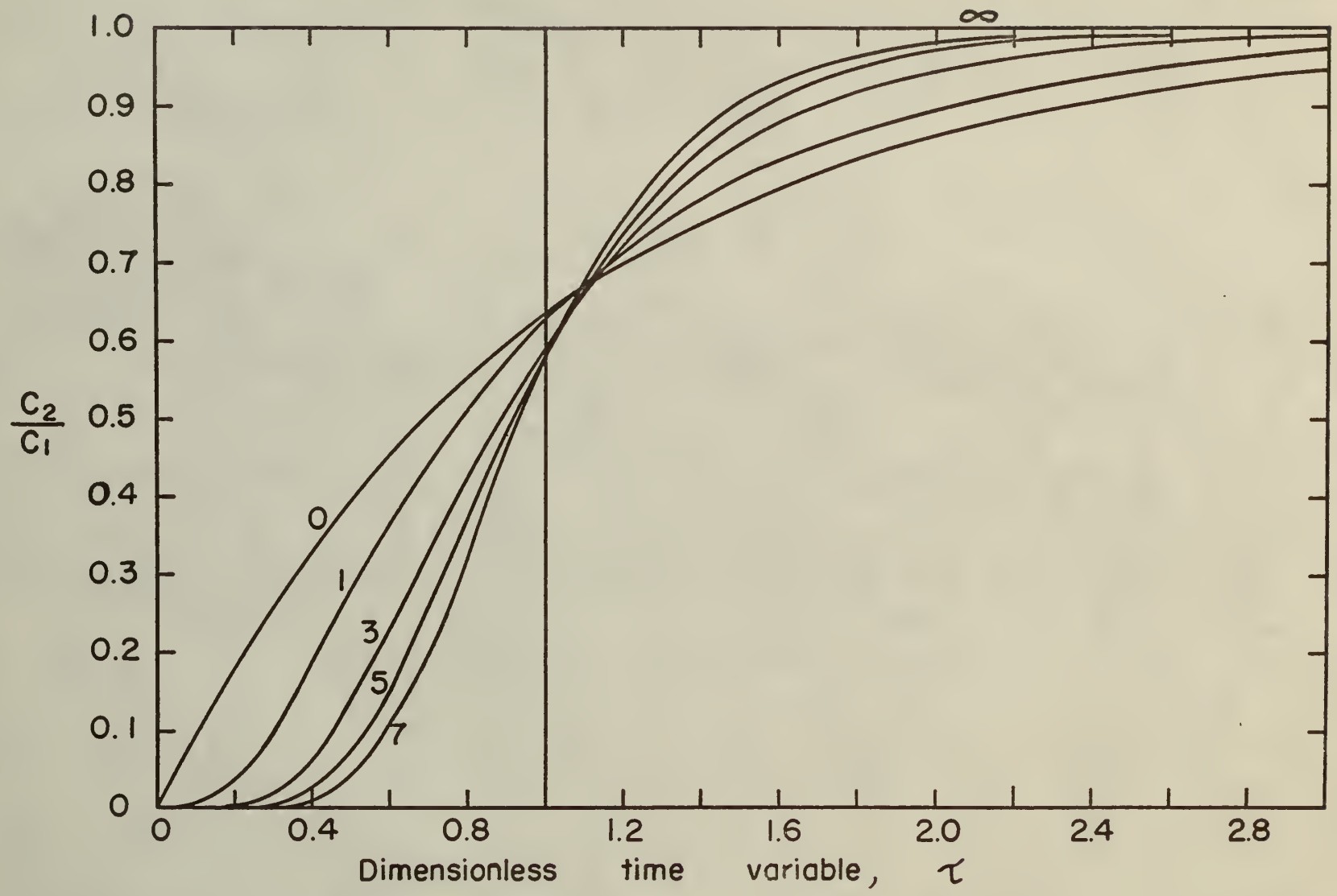

Fig. 22. Calculated response curves of exit concentration against step input for non-reacting systems (R=0) based on B. C. (I) with M as parameter.

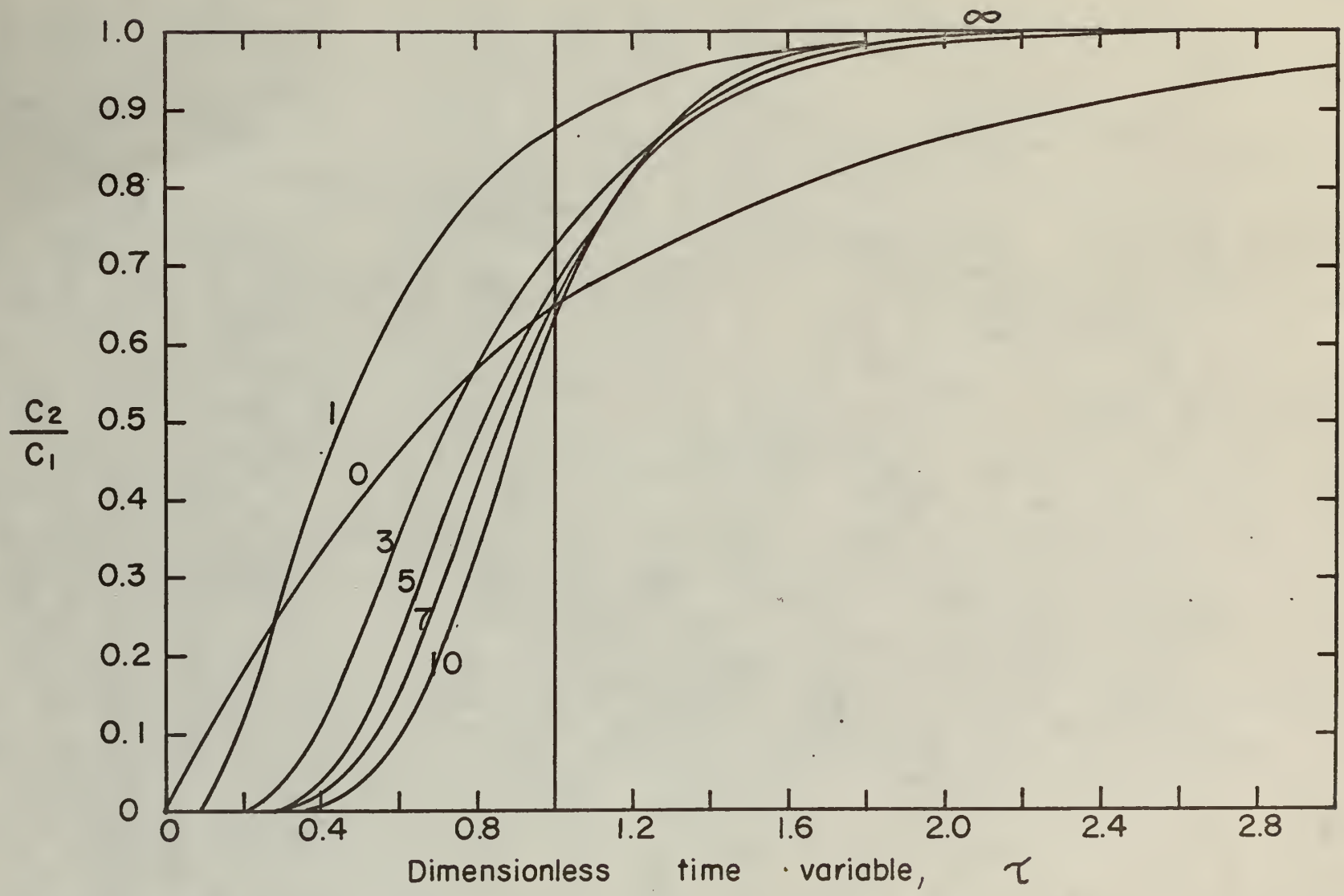

Fig. 23. Calculated response curves of exit concentration against step input for non reacting systems (R=0) based on B. C. (II) with M as parameter.

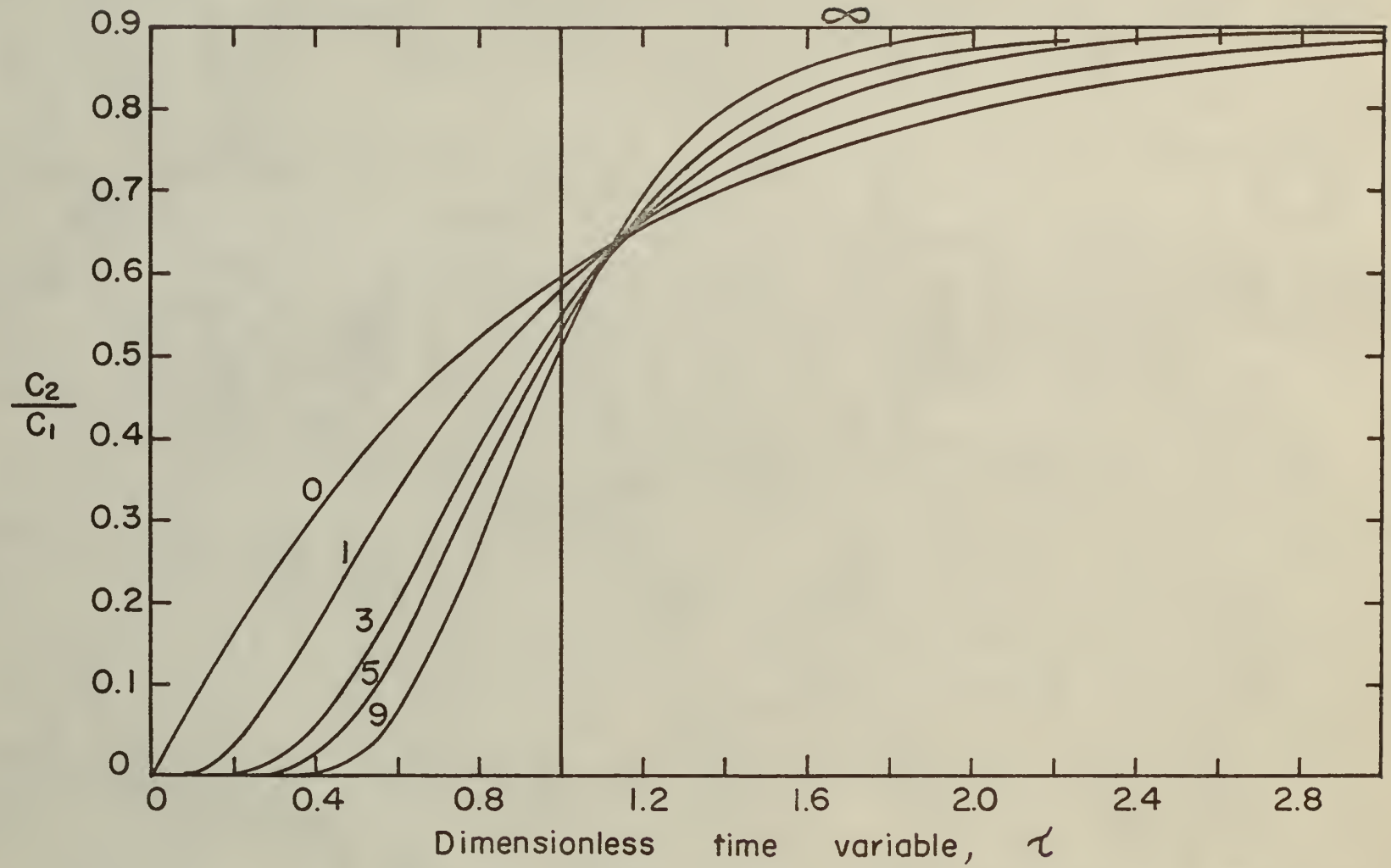

Fig. 24. Calculated response curves of exit concentration against step input for reacting systems (R=0.1) based on B. C. (I) with M as parameter.

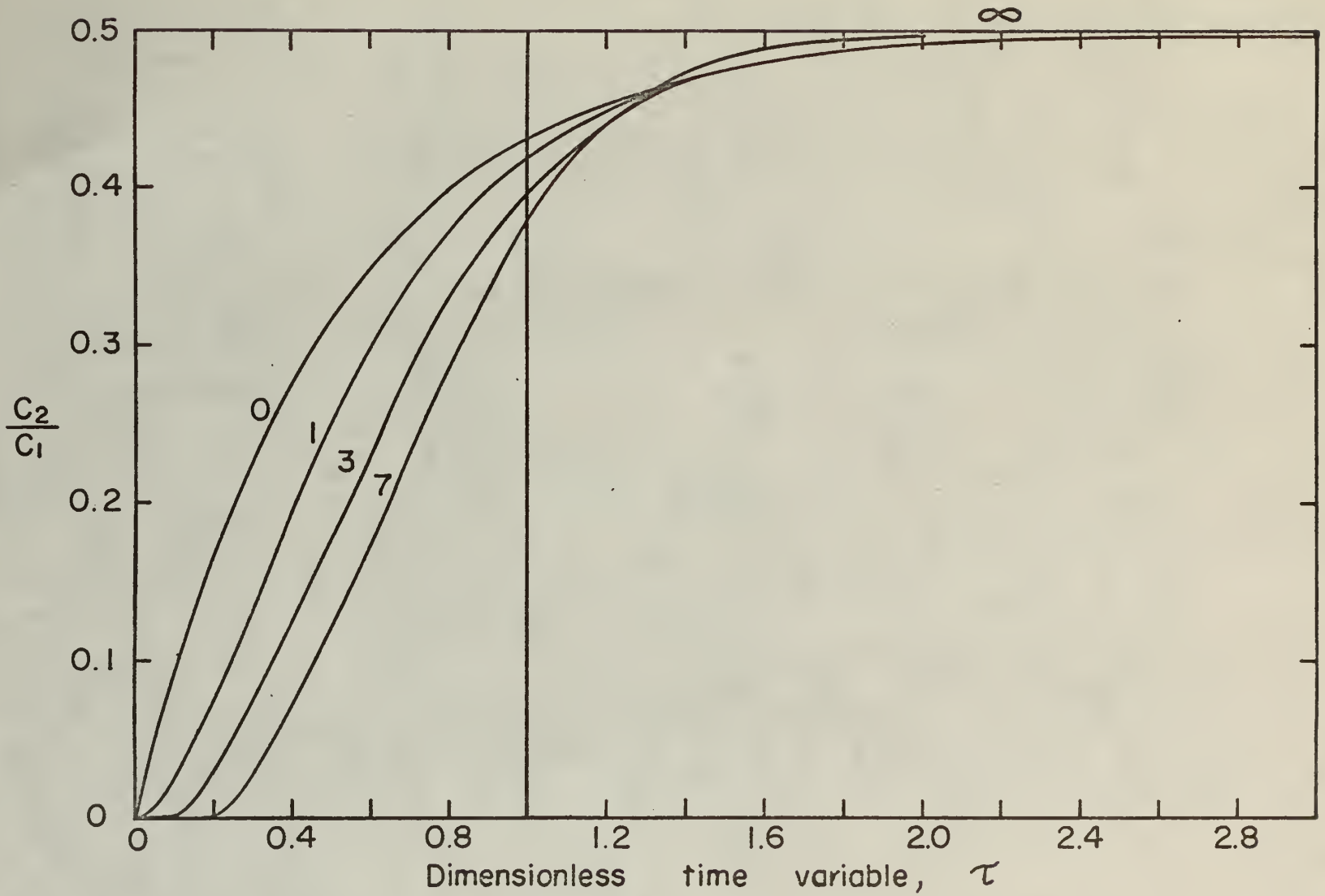

Fig. 25. Calculated response curves of exit concentration against step input for reacting systems (R=1) based on B. C. (I) with M as parameter.

The results of frequency response curves based on B.C.(I) (equation 31) are shown in Figs. 26 through 28 in the form of dimensionless Bode plots. Frequency responses of the two limiting conditions of the dispersion model based on equations (38) and (43) are also shown in the figures for the sake of comparison. Only the frequency response curves based on B.C.(I) are presented herewith because of the consistency observed in the previous section with the two limiting cases. Other frequency curves based on different sets of boundary conditions are presented only for the sake of comparison in Figs. 29 through 34.

Figure 26 shows that the plots of amplitude ratios and phase shifts change monotonically between the two limits for the nonreacting systems in the range of $\omega\theta$ which is of practical interest (15). This, however, is no longer true for the reacting systems, as shown in Figs. 27 and 28. As the relative reaction rates characterized by R increase, amplitude ratio decreases rapidly, especially at large values of M. For example, at the conditions for which R and M are greater than 6 and 10 respectively, the amplitude ratio is very small and practically is negligible. Under these conditions of rapid reaction and small degree of dispersion, a cyclic unsteady-state system has little effect on the performance of the system; and therefore, the frequency response data are of little use. In contrast, the phase shift data are only slightly affected by the change in the values of M and R. Thus, it can be said that the phase shift data are of little value in determining the characteristics of the system.

The performance function represented by equation (118) (See

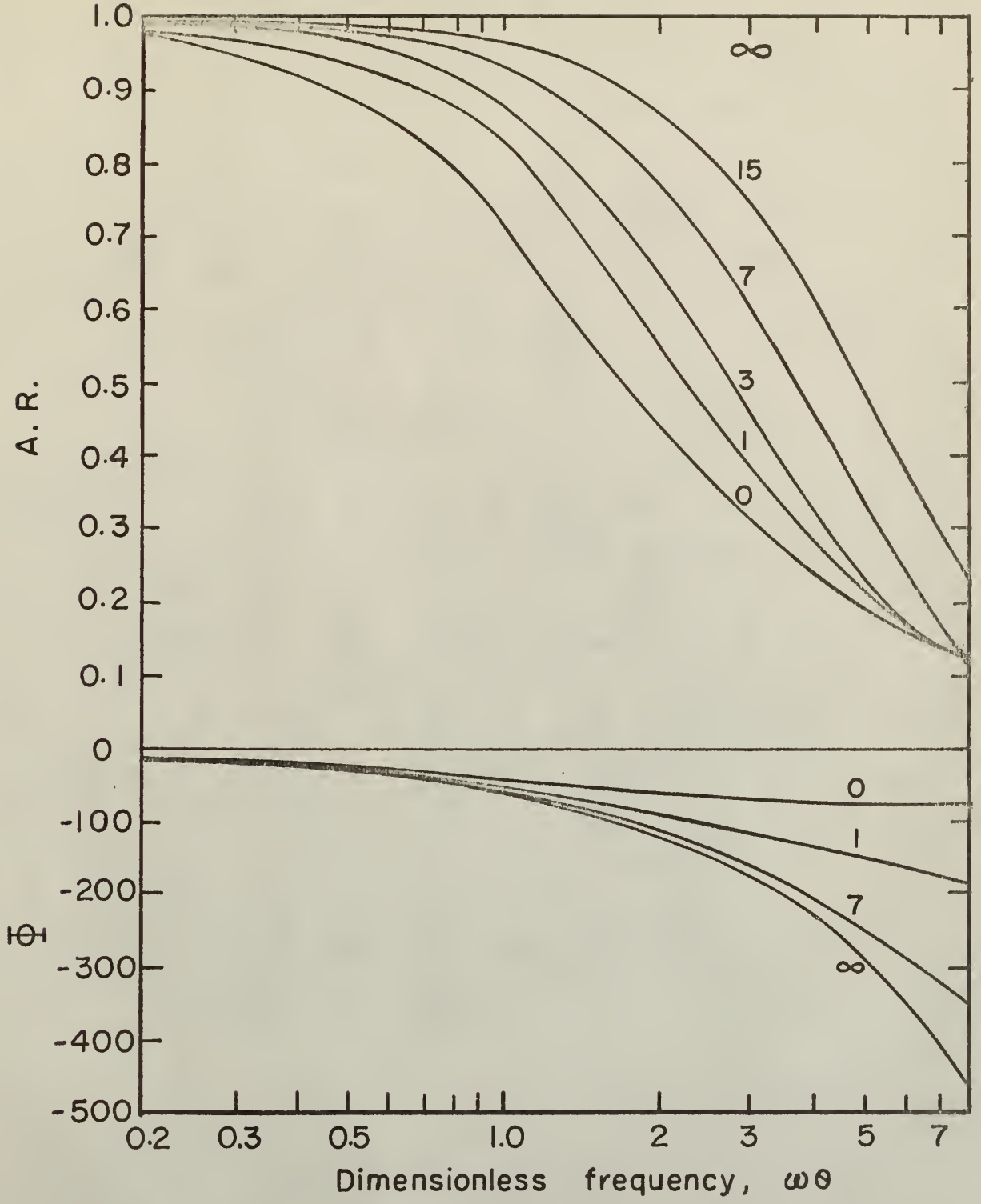

Fig. 26. Frequency response diagram of non-reacting system (R=0) based on B.C. (I) with M as parameter.

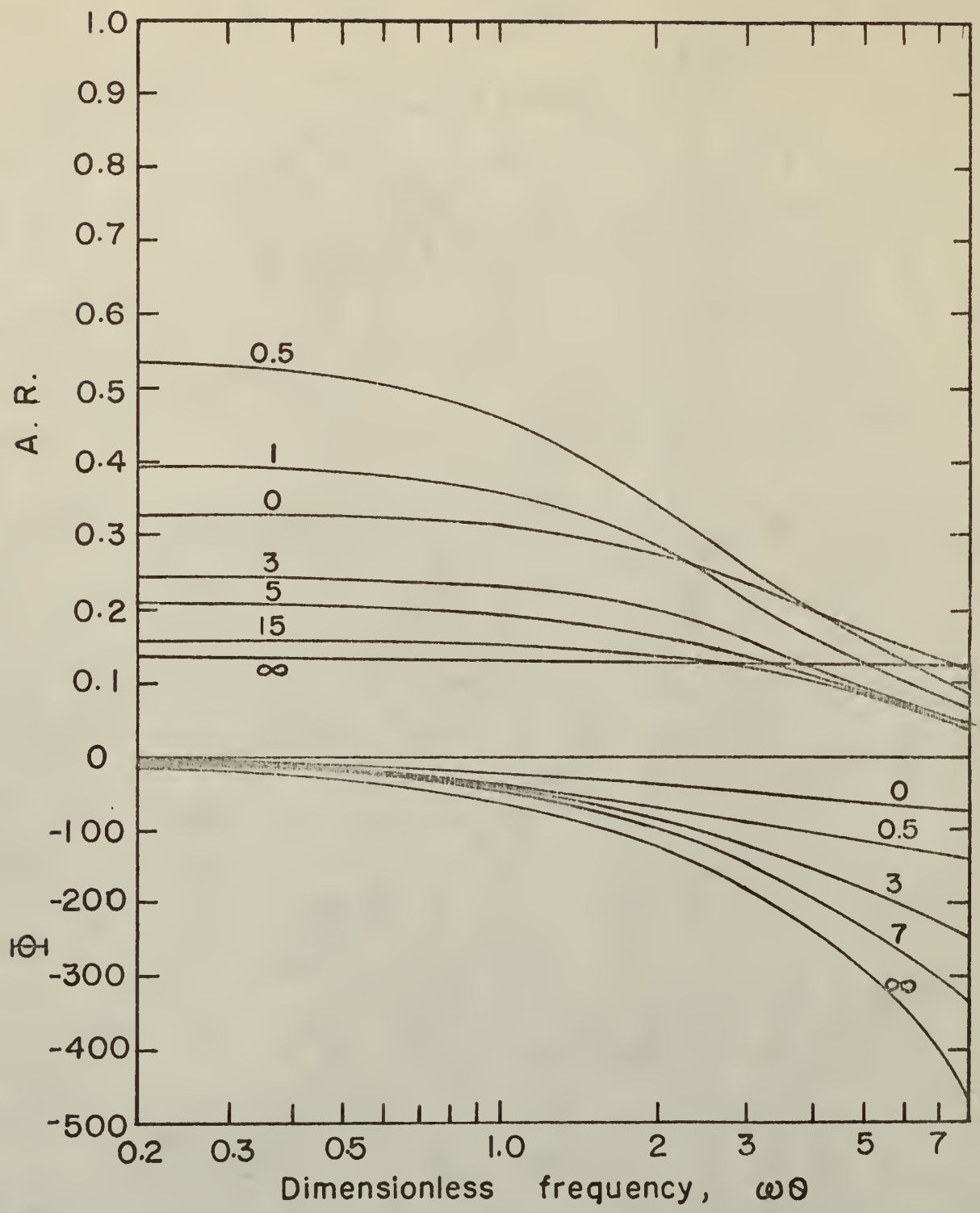

Fig. 27. Frequency response diagram of reacting system (R=2) based on B. C. (I) with M as parameter.

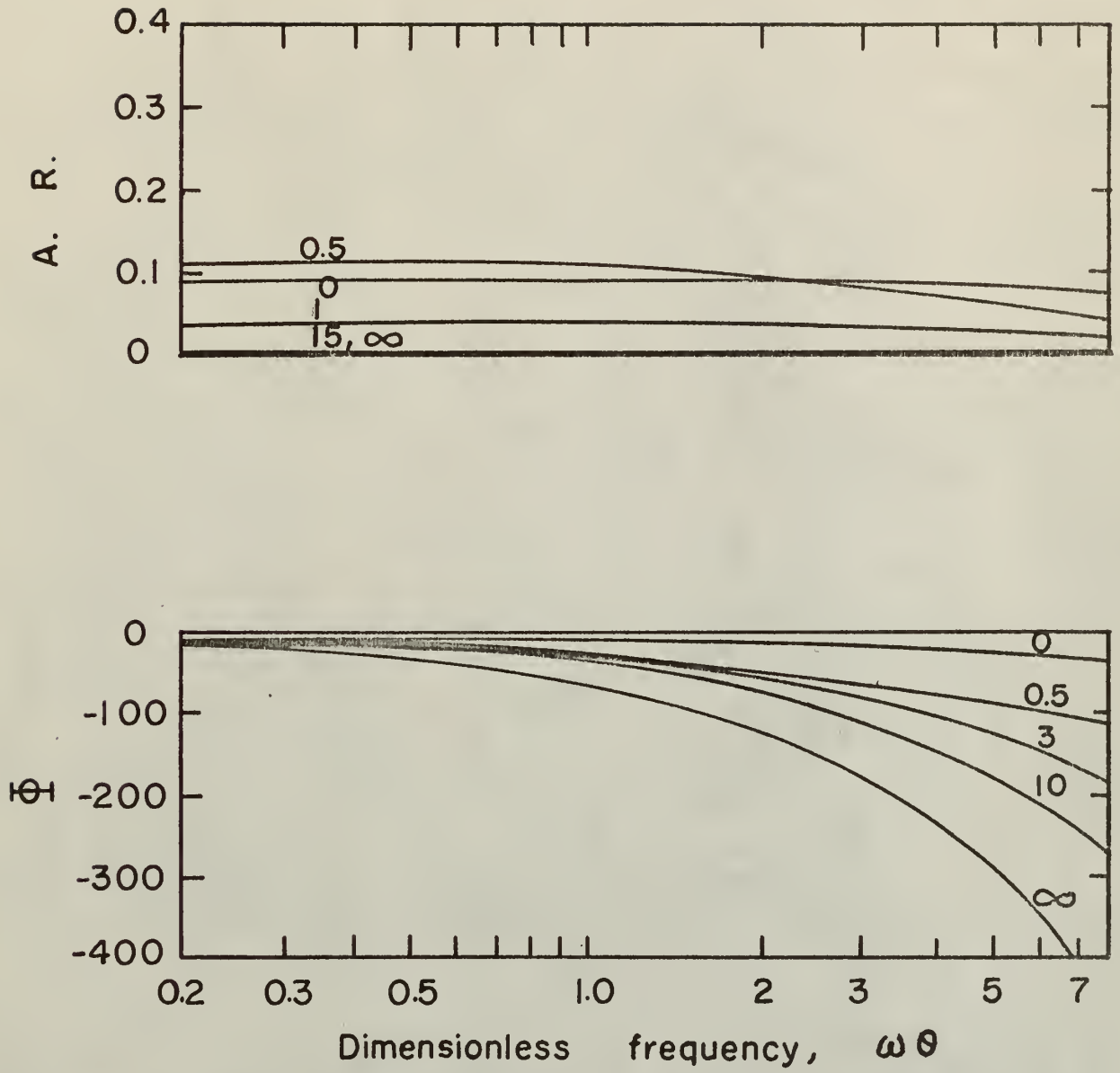

Fig. 28. Frequency response diagram of reacting system (R=10) based on B. C. (I) with M as parameter.

Appendix IV) was originally derived by Kramers and Alberda (10) for non-reacting systems. In order to avoid complexities in transforming equation (118) to the computable form of equation (31), it was suggested that equation (31) be approximated by the performance function of n numbers of the perfectly mixed vessels connected in series. Such approximation is good only for large values of M(c.a. M = 15). Very frequently approximate boundary conditions which do not conserve the continuity requirements, i.e., B.C.(II) and (III), are employed. Kramers and his coworkers (11) employed B.C.(II) and (III), and the evaluation of the performance function based on B.C.(III) (equation 108 of Appendix IV) has been frequently presented in the literature (7) (22).

The amplitude ratios and phase shift based on these different sets of boundary conditions are compared in Figs. 29 through 34 both with and without chemical reaction at high and low degree of dispersion. As can be seen from Fig. 29, the agreement of the amplitude ratios and phase shift curves for non-reacting systems based on approximate boundary conditions are quite poor at low values of M (large degree of dispersion). An increasing degree of agreement, however, is observed in Fig. 30 as M increases. The deviations among the phase shift curves are very small when M is approximately 10 or greater, and the amplitude ratio curves practically coincide when M is approximately 15 or greater. It is also seen that phase shift curves are generally less affected than amplitude ratio curves. The performance function curves of the reacting system, shown in Figs. 31 through 34, reveal the effects of using different sets of boundary conditions more clearly than

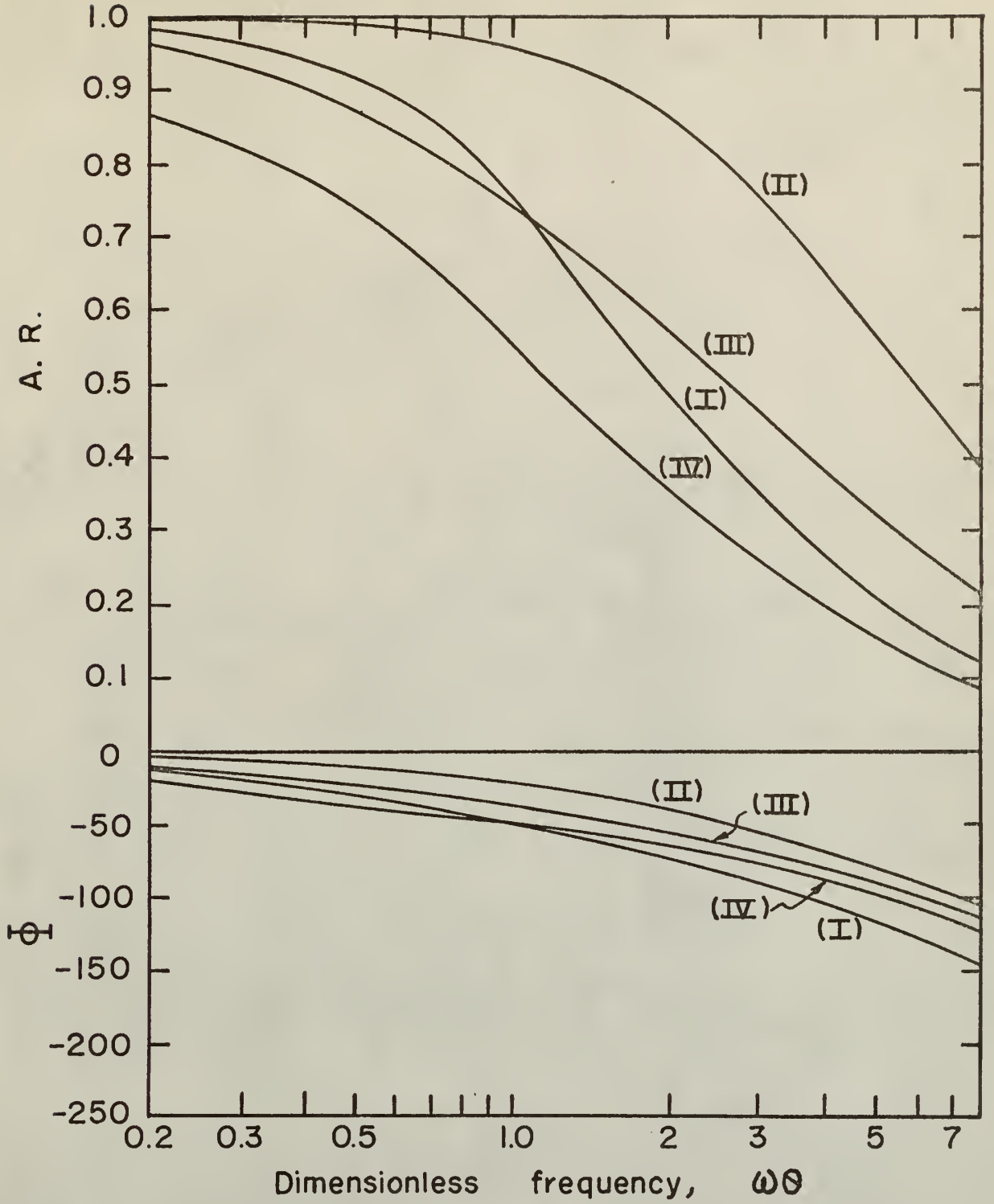

Fig. 29. Comparison of frequency response diagrams of non-reacting system (R=0) based on different boundary conditions at high degree of dispersion, $M=\frac{1}{2}$.

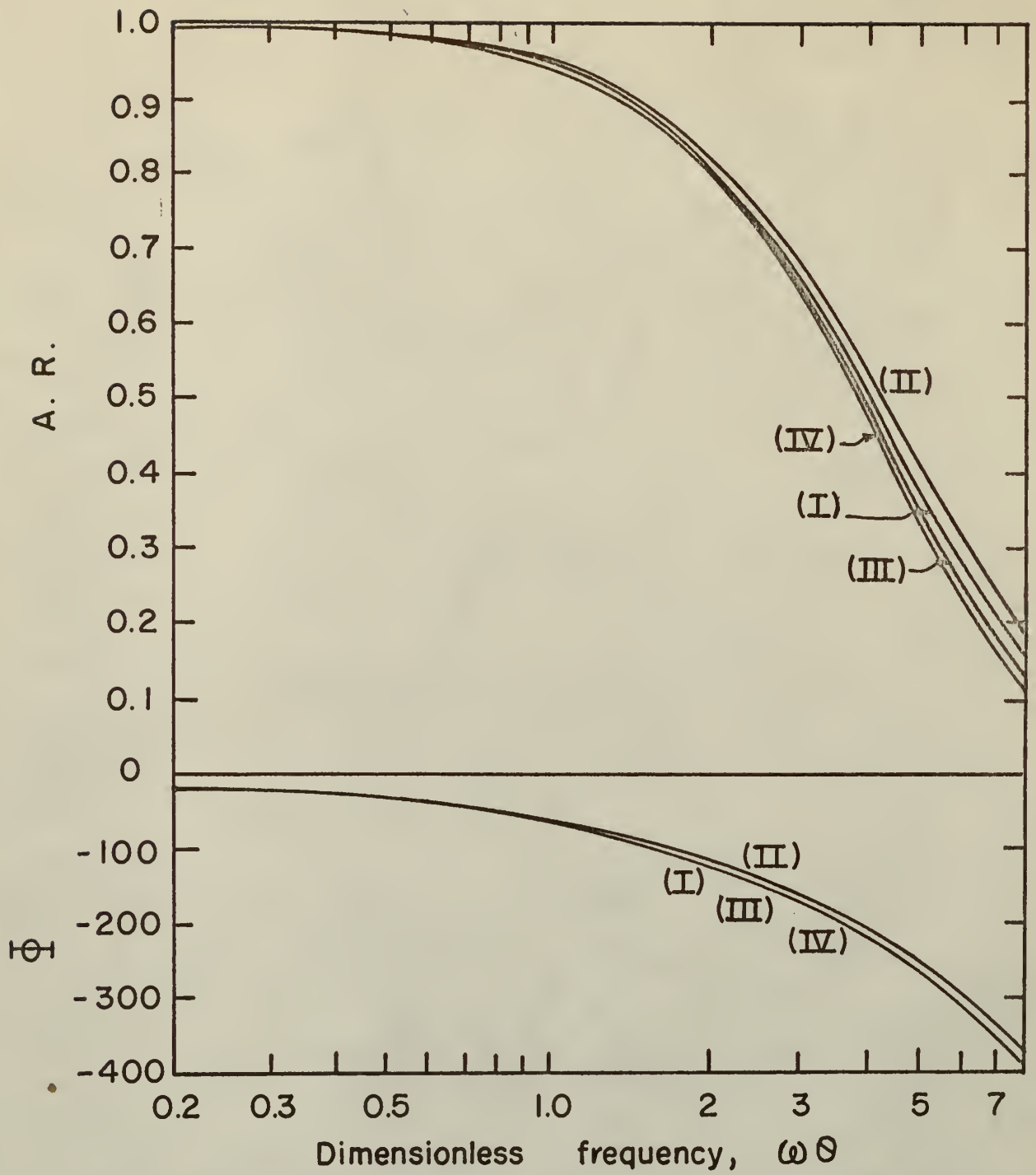

Fig. 30. Comparison of frequency response diagrams of non-reacting system (R=0) based on different boundary conditions at low degree of dispersion, M=10.

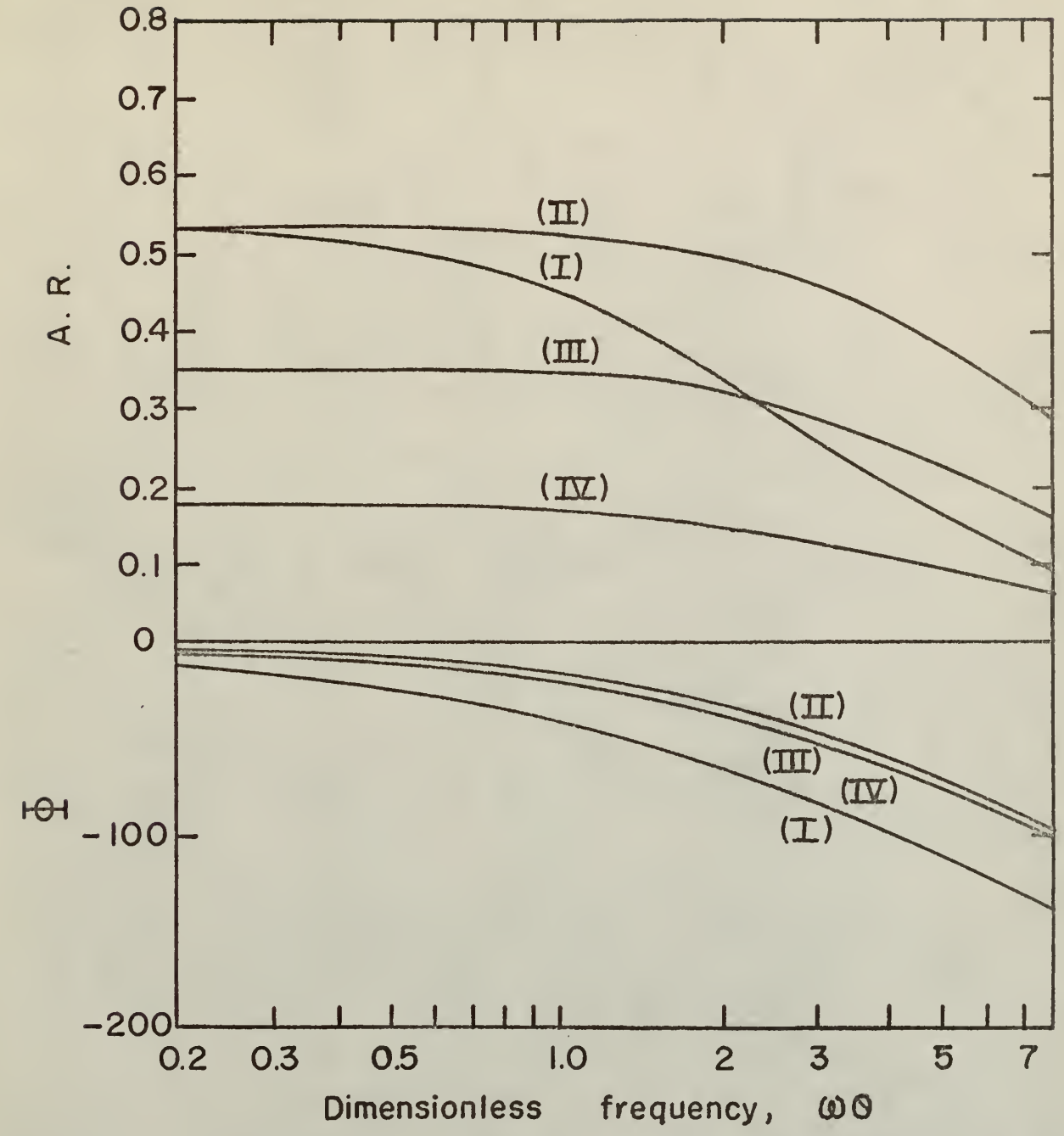

Fig. 31. Comparison of frequency response diagrams of reacting system (R=2) based on different boundary conditions at high degree of dispersion, $M = \frac{1}{2}$.

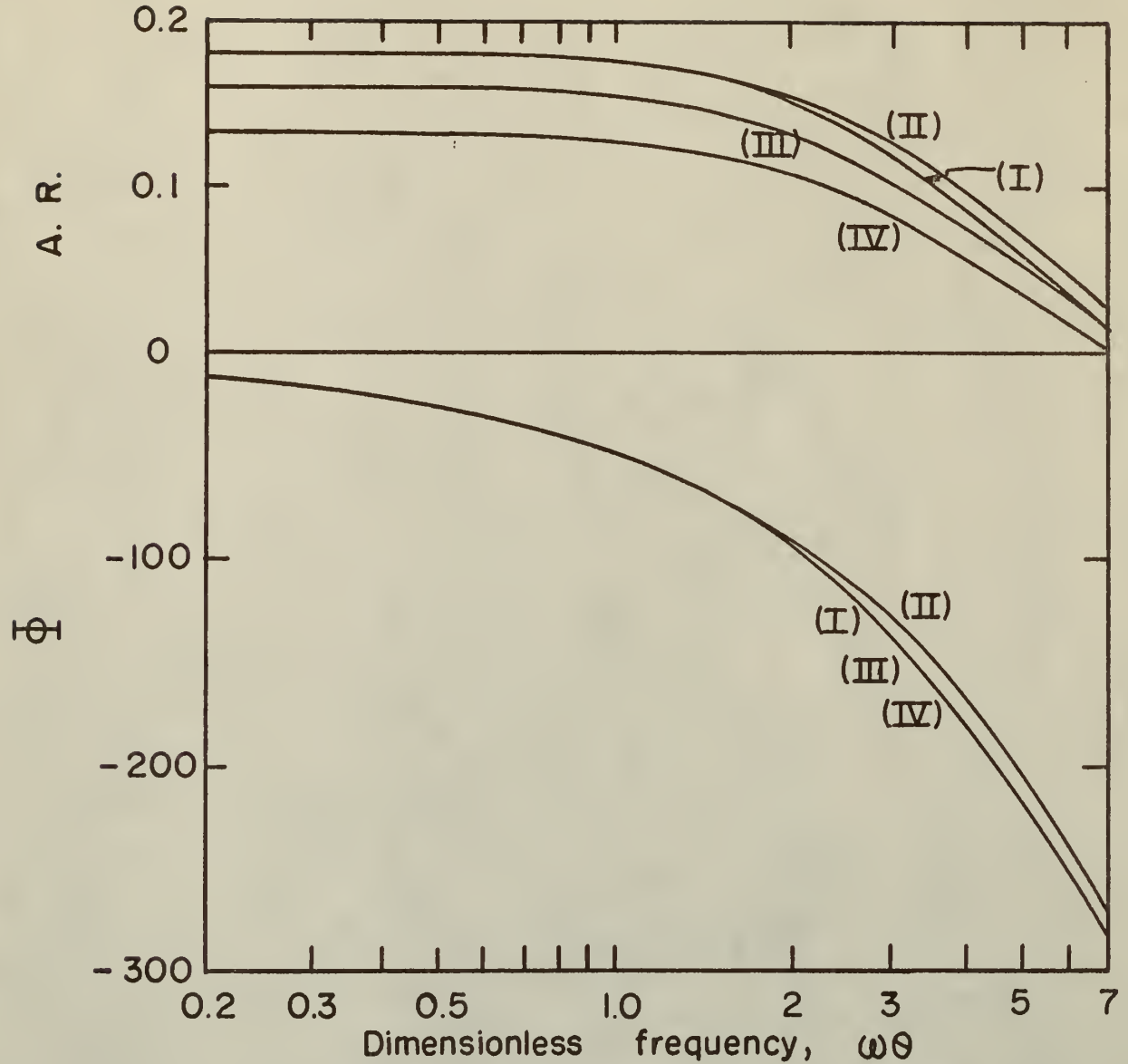

Fig. 32. Comparison of frequency response diagrams of reacting system (R=2) based on different boundary conditions at low degree of dispersion, M = 7.

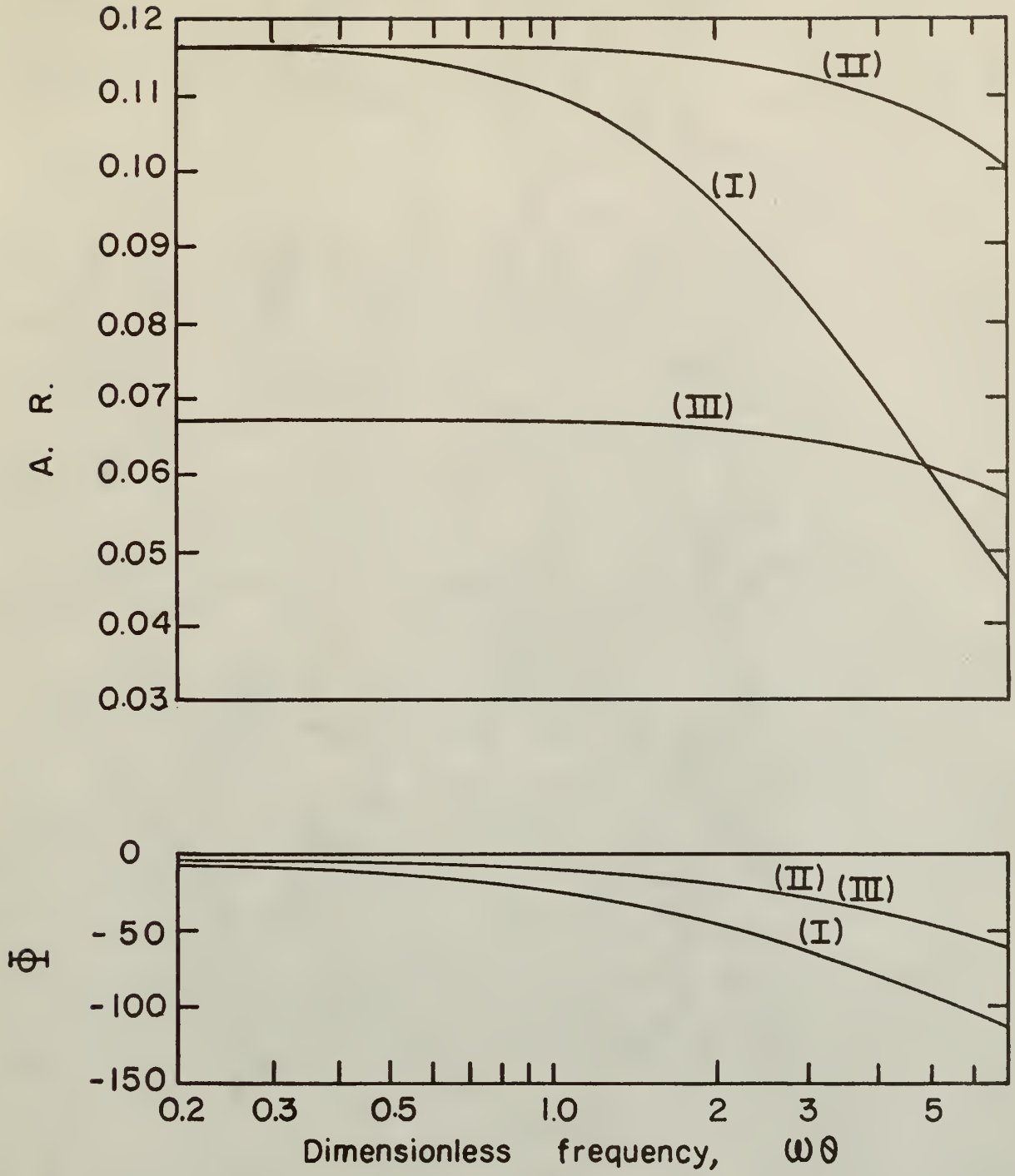

Fig. 33. Comparison of frequency response diagrams of reacting system (R=10) based on different boundary conditions at high degree of dispersion, $M = \frac{1}{2}$.

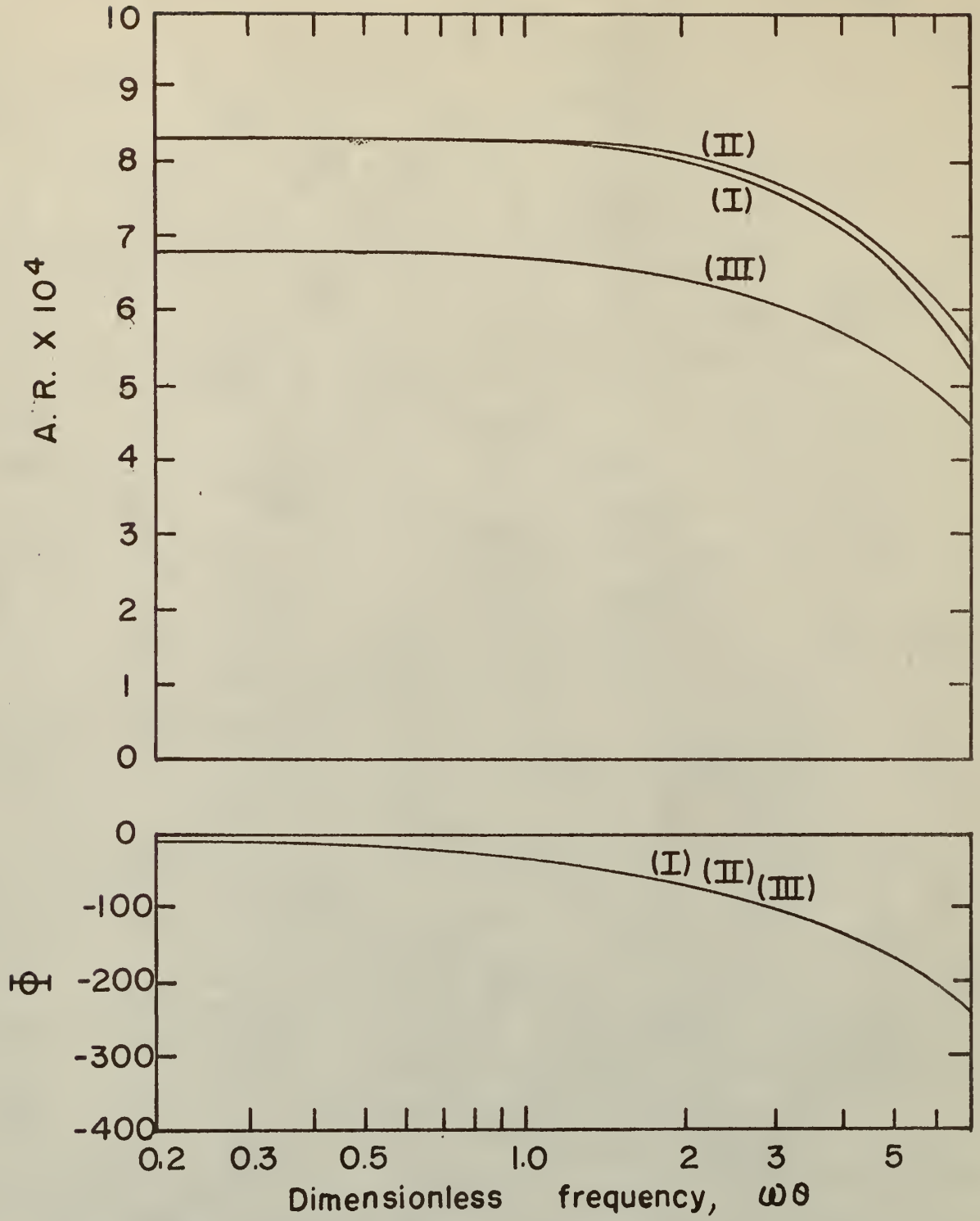

Fig. 34. Comparison of frequency response diagrams of reacting system (R=10) based on different boundary conditions at low degree of dispersion, M=10.

the corresponding curves for the non-reacting systems -- the greater the value of R, the greater the effect of the boundary conditions. It is also seen from these figures that, at any given relative reaction rate characterized by R, the effect of different sets of boundary conditions becomes smaller as the degree of mixing decreases.

An illustration of the determination of the mean dispersion coefficient from a frequency response experiment for non-reacting systems by comparing available experimental data (7) (10) (22) with calculated values is presented in Figs. 35 through 37. These figures show that the use of approximate boundary conditions can give rise to an appreciable error in determining the degree of dispersion, especially at low values of M. As has been discussed in the previous section, the use of B.C.(III) is quite desirable under some circumstances due to the simplicity of equation (33). For the preliminary design of a tubular flow system and the synthesis of a complex control system in which the use of a complicated performance function is involved, the use of approximate boundary conditions should be avoided.

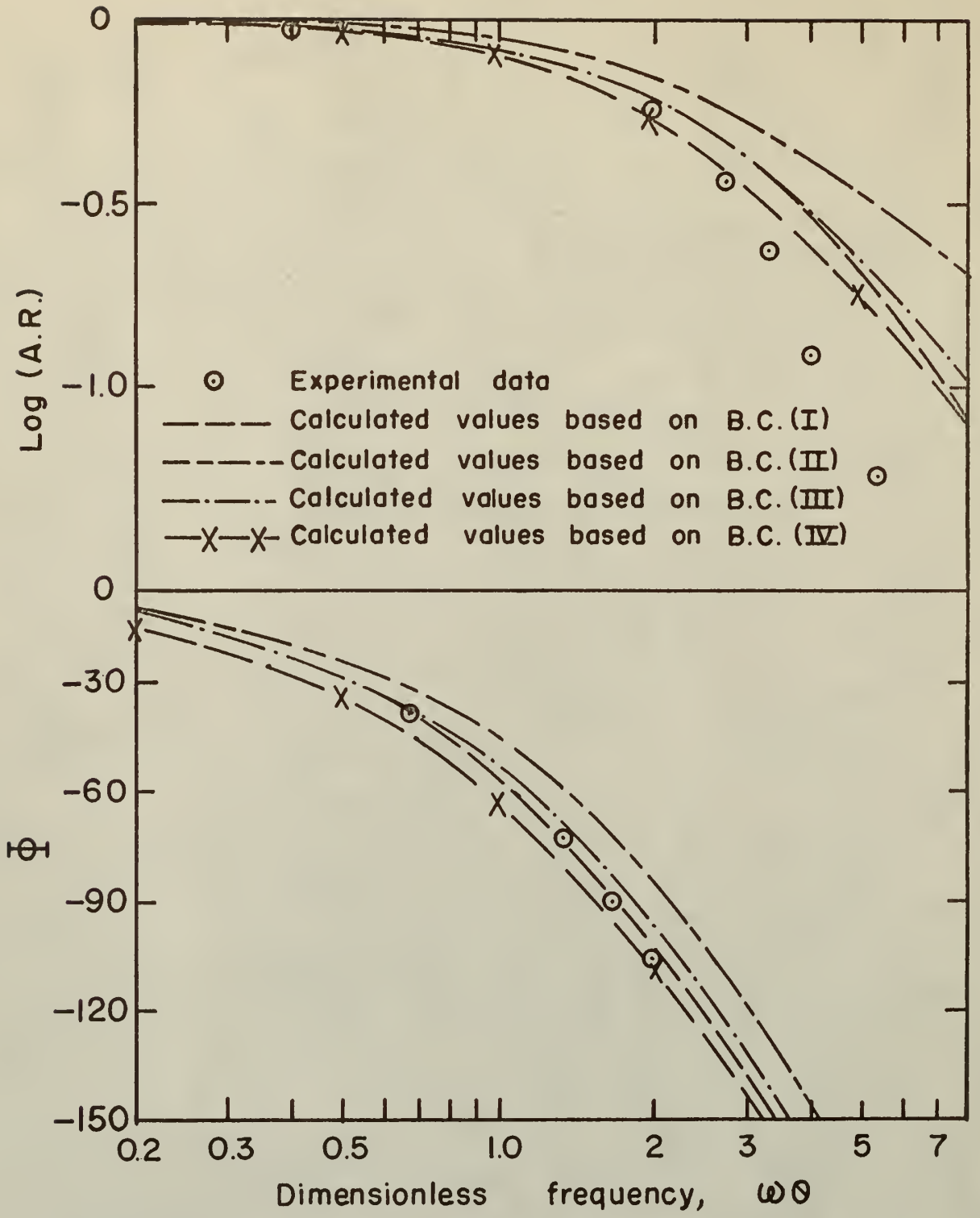

Fig. 35. Comparison of experimental and calculated frequency response diagrams.

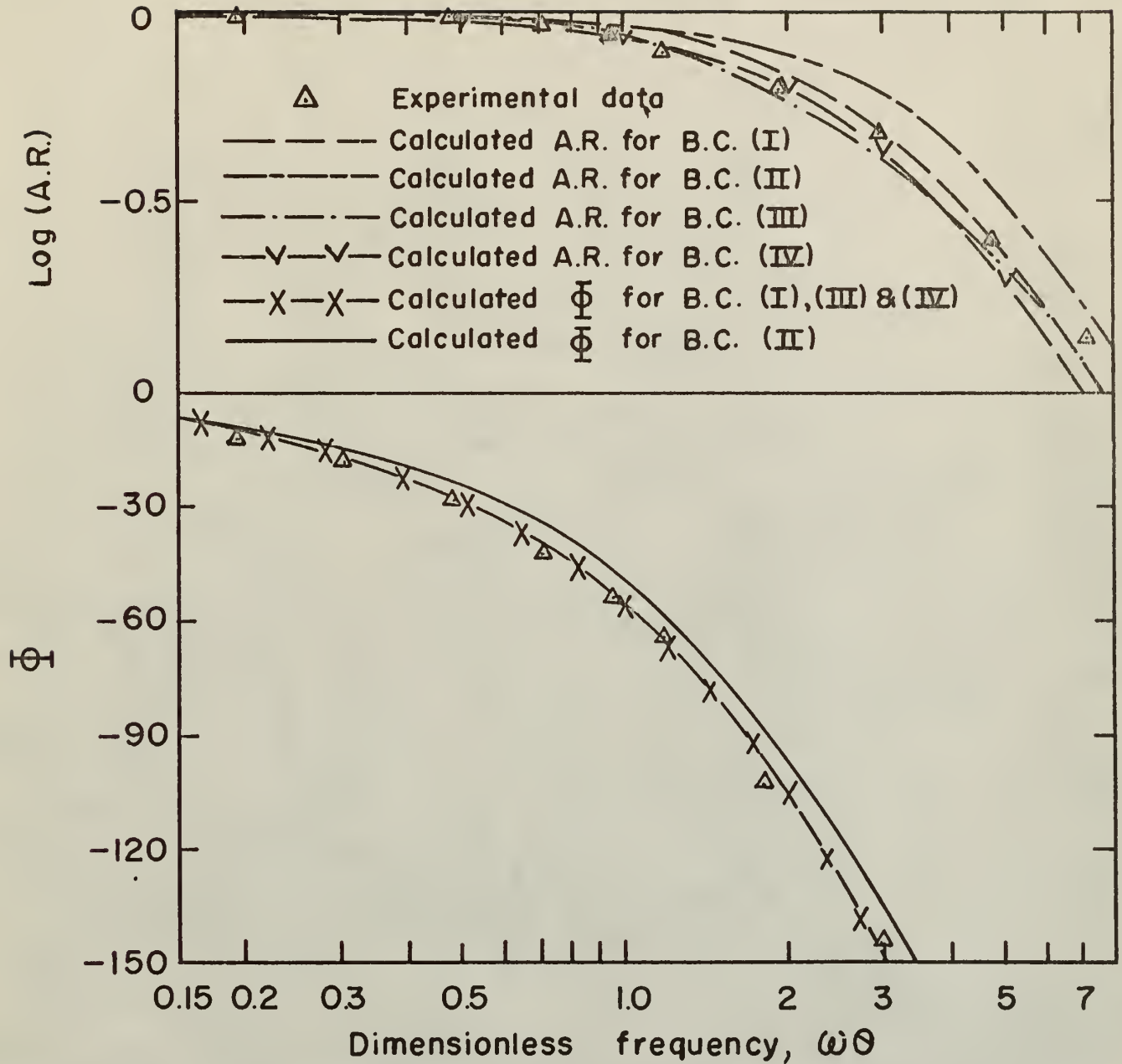

Fig. 36. Comparison of experimental and calculated frequency response diagrams.

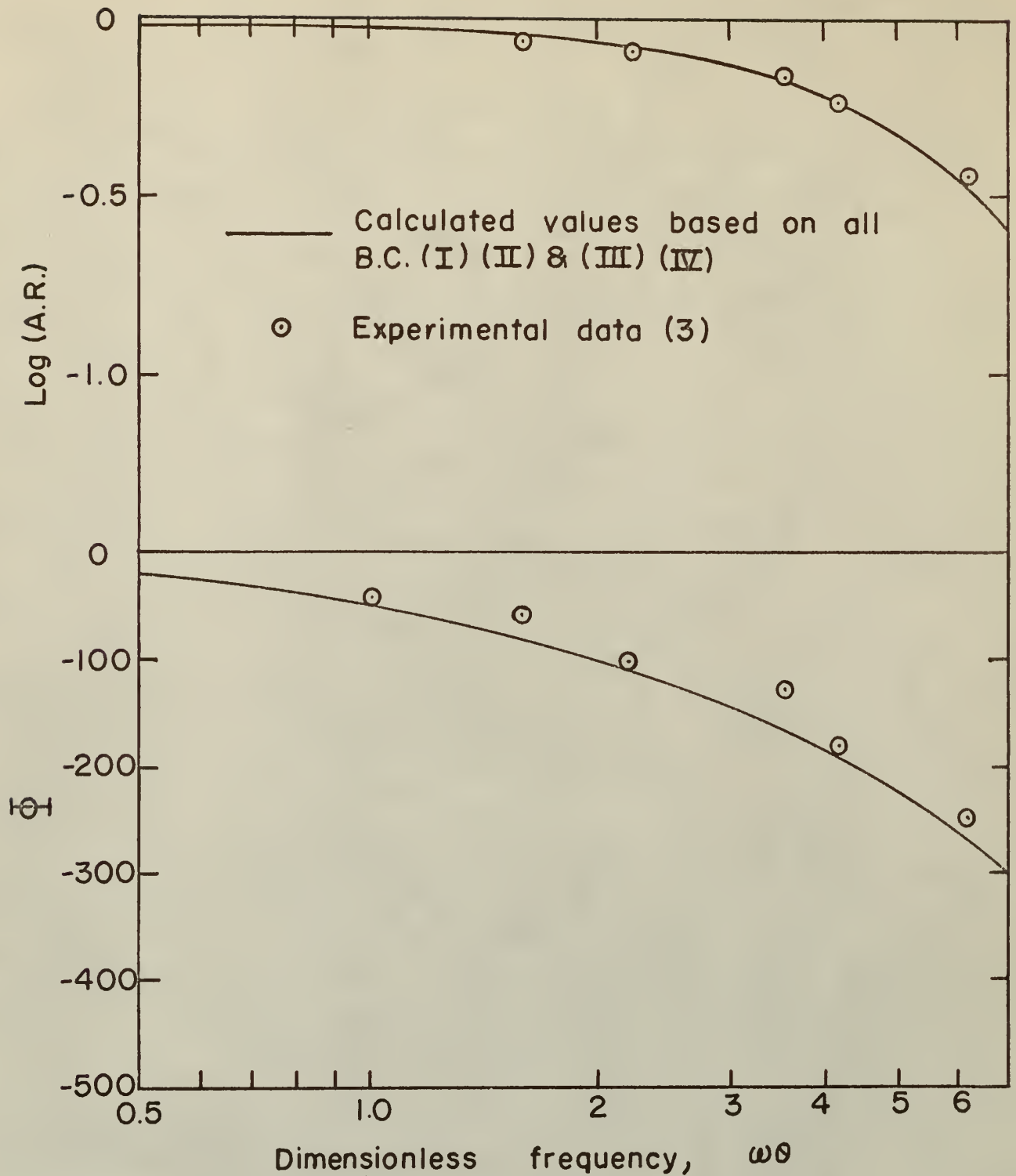

Fig. 37. Comparison of experimental and calculated frequency response diagrams.

CONCLUSION

The general extent of axial dispersion and its effect on the performance of isothermal tubular flow reactors can be characterized by a diffusion-type model with appropriate boundary conditions. Among the four boundary conditions considered, only B.C.(I) yields conversions and concentration profiles consistent with the theoretical limits of both a uniformly stirred tank reactor and a plug flow type reactor, and for this reason, the use of B.C.(II), (III) and (IV) should be avoided in and around the domain of inconsistent conversions based on a one-dimensional transport equation.

There are no data available on transient response experiments accompanied by first-order chemical reaction. If the data were available, the calculated response curves based on first-order chemical reaction could be used to determine the dispersion coefficient by the same method presented in this research for the non-reacting case.

The response curves presented in this research for various types of transient input should be useful for practicing process control engineers and for those who wish to extract the transport parameters from dynamic response experiments.

ACKNOWLEDGMENT

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NOMENCLATURE

a = angle defined as $\frac{1}{2}\,\mathrm{Tan}^{-1}\left[\frac{2(\omega\theta)}{2R+M}\right]$, degree.

A = a constant in equation (27) which is numerically equal to the steady-state conversion, dimensionless.

(A.R.) = amplitude ratio, dimensionless.

b = $\left[\left(1+\frac{2R}{M}\right)^2+\frac{4(\omega\theta)^2}{M^2}\right]^{\frac{1}{4}}$

B = a constant in equation (28) which is numerically equal to the steady-state conversion, dimensionless.

C = concentration of reactant, m/l.

$C_1(t)$ = inlet concentration of reactant, m/l.

$C_2(t)$ = exit concentration of reactant, m/l.

D = axial dispersion coefficient, l^2/t.

$\bar{D}$ = mean value of D, l^2/t.

f_i = conversion calculated based on B.C.(i), dimensionless.

$f_{M=0}$ = conversion calculated based on a stirred tank reactor, dimensionless.

$f_{M=\infty}$ = conversion calculated based on a plug flow reactor, dimensionless.

j = $\sqrt{-1}$

k = reaction rate constant, t^{-1}.

L = length of reactor, l.

M = $\bar{u}L/2\bar{D}$, 1/2 axial of Peclet number, dimensionless.

n = order of reaction.

P.F. = performance function defined as the ratio of Fourier transform of output to input.

Q = amount of tracer injected in impulse function, m.

R = $kL/\bar{u}$, reaction number, dimensionless.

$r(C)$ = rate of reaction.

s = function of Laplace transform.

t = time.

$\bar{u}$ = average flow velocity of reacting mixture, l/t.

v = volumetric flow rate, l^3/t.

V = volume of reactor, l^3.

$W(s)$ = transfer function defined as the Laplace transform of output to input.

x = Mb cos a

y = Mb sin a

z = co-ordinate in the direction of flow, l.

Greek Letters

$$\alpha_1 = \sinh x \cos y - \frac{(\omega\theta)}{M} \cosh x \sin y + b(\cos a \cosh x \cos y - \sin a \sinh x \sin y).$$

$$\alpha_2 = \left(\frac{\omega\theta}{M}\right) \sinh x \cos y + \cosh x \sin y + b(\cos a \sinh x \sin y + \sin a \cosh x \cos y).$$

$$\alpha_3 = \sinh x \cos y + b(\cos a \cosh x \cos y - \sin a \sinh x \sin y).$$

$$\alpha_4 = \cosh x \sin y + b(\sin a \cosh x \cos y + \cos a \sinh x \sin y).$$

$$\beta = (1 + 2R/M)^{\frac{1}{2}}.$$

$$\beta' = \left[1 + (2\theta/M)(s + k)\right]^{\frac{1}{2}}$$

$$\beta'' = \left[1 + (2\theta/M)(j\omega + k)\right]^{\frac{1}{2}}$$

δ_n = roots of $\cot \delta = \frac{1}{2}\left(\frac{\delta}{M} - \frac{M}{\delta}\right)$.

δ'_n = roots of $\tan \delta' = -\frac{\delta'}{M}$.

ϕ = phase shift, degree.

η = z/L, dimensionless distance from the inlet of reactor.

γ = C/C_1, dimensionless concentration.

τ = the dimensionless time variable, t/θ .

τ_0 = thickness of impulse in time domain, dimensionless.

θ = $L/\bar{u}$, t.

ω = angular frequency, rad./t.

APPENDIX

APPENDIX I

Derivations of Steady-state Solutions

The solutions of equation (8) which satisfy the boundary conditions (I), (II), (III) and (IV) are obtained by means of Laplace transformations.

Laplace transform of equation (8) with respect to η is:

$$\gamma(s) = \frac{s\,\gamma(0^+) + \gamma'(0^+) - 2M\,\gamma(0^+)}{s^2 - 2Ms - 2MR} \tag{48}$$

For the solution using B.C.(I), the Laplace transform of equation (3) is substitued in equation (48) to obtain:

$$\gamma(s) = \frac{s\,\gamma(0^+) - 2M}{s^2 - 2Ms - 2MR} \tag{49}$$

By means of partial fractionation, equation (49) is separated into two parts:

$$\gamma(s) = \frac{(1+\beta)\,\gamma(0^+) - 2}{2\beta[s - M(1+\beta)]} - \frac{(1-\beta)\,\gamma(0^+) - 2}{2\beta[s - M(1-\beta)]} \tag{50}$$

The inverse transform of equation (50) gives:

$$\gamma(\eta) = \frac{(1+\beta)\,\gamma(0^+) - 2}{2\beta} \exp\left[M(1+\beta)\eta\right]$$

$$- \frac{(1-\beta)\,\gamma(0^+) - 2}{2} \exp\left[M(1-\beta)\eta\right] \tag{51}$$

According to boundary condition represented by equation (4), differentiation of equation (51) and setting it equal to zero

gives:

$$\gamma(0^+) = \frac{2\{(1+\beta)\exp[M(1+\beta)] - (1-\beta)\exp[M(1-\beta)]\}}{(1+\beta)^2\exp[M(1+\beta)] - (1-\beta)^2\exp[M(1-\beta)]} \qquad (52)$$

Substitution of equation (52) into (51) and simplification gives equation (9) of text.

For the solution using B.C.(II), the Laplace transform of equation (5) is substituted into equation (48) and simplified to:

$$\gamma(s) = \frac{(s - 2M) + \gamma(0^+)}{s^2 - 2Ms - 2MR} \qquad (53)$$

By means of partial fractionation, equation (53) is separated into two parts as:

$$\gamma(s) = \frac{-M(1-\beta) + \gamma'(0^+)}{2M\beta[s-M(1+\beta)]} - \frac{-M(1+\beta) + \gamma'(0^+)}{2M\beta[s-M(1-\beta)]} \qquad (54)$$

The inverse transform of equation (54) yields:

$$\gamma(\eta) = \frac{-M(1-\beta) + \gamma(0^+)}{2M\beta}\exp[M(1+\beta)\eta] - \frac{-M(1+\beta) + \gamma'(0^+)}{2M\beta}\exp[M(1-\beta)\eta] \qquad (55)$$

According to boundary condition represented by equation (4), equation (55) is differentiated in order to solve for $\gamma'(0^+)$. It gives:

$$\gamma'(0^+) = M(1-\beta^2)\left\{\frac{\exp[M(1+\beta)] - \exp[M(1-\beta)]}{(1+\beta)\exp[M(1+\beta)] - (1-\beta)\exp[M(1-\beta)]}\right\} \qquad (56)$$

Substitution of equation (56) into (55) and simplification gives equation (10) of text.

For the solution using B.C.(III), substitution of equation (5) into equation (48) gives equation (55). As η approaches infinity in equation (55), the second term in the right-hand side of equation (55) vanishes as:

$$\frac{M(\beta - 1) + \gamma(0^+)}{2M\beta} = 0 \tag{57}$$

Solving for $\gamma(0^+)$ in equation (57) and substituting it into equation (55), equation (11) of text is obtained.

For the solution using B.C.(IV), substitution of equation (3) into equation (48) gives equation (51). As η approaches infinity in equation (51), the first term in the right-hand side of equation (51) vanishes, hence

$$0 = \frac{(1+\beta)\ \gamma(0^+) - 2}{2\beta} \tag{58}$$

Solving for $\gamma(0^+)$ in equation (58) and substituting it into equation (51) yields equation (12) of text.

APPENDIX II

Derivations of the Transfer Functions, W(s)

The Laplace transform of equation (2) with respect to t at zero initial concentration is:

$$sC(z,s) = \bar{D}\frac{\partial^2 C(z,s)}{\partial z^2} - \bar{u}\frac{\partial C(z,s)}{\partial z} - kC(z,s) \tag{59}$$

Equation (59) is in the form of a second-order differential equation whose general solution is:

$$C(z,s) = A_1 e^{\frac{\bar{u}z}{2\bar{D}}(1+\beta)} + A_2 e^{\frac{\bar{u}z}{2\bar{D}}(1-\beta)} \tag{60}$$

where A_1, A_2 = arbitrary constants.

The transfer functions, defined as the ratio of the Laplace transforms of output to input, then become

$$W(s) = \frac{C(L,s)}{C_1(0,s)} \tag{61}$$

In order to solve for the arbitrary constants in equation (60), the boundary conditions tabulated in Table 2 are transformed as:

B.C.(I)

$$\bar{u}C(0,s) = \bar{u}C(0^+,s) - \bar{D}\frac{dC(0^+,s)}{dz} \tag{62a}$$

$$\frac{\partial C(L,s)}{\partial z} = 0 \tag{62b}$$

B.C.(II)

$$\bar{u}C(0,s) = \bar{u}C(0^+s) \tag{63a}$$

$$\frac{\partial C(L,s)}{\partial z} = 0 \tag{63b}$$

B.C.(III)

$$\bar{u}C(0,s) = \bar{u}C(0^+,s) \tag{64a}$$

$$\lim_{z\to\infty} C(z,s) = 0 \tag{64b}$$

B.C.(IV)

$$\bar{u}C(0,s) = \bar{u}C(0^+,s) - \bar{D}\frac{\partial C(0^+,s)}{\partial z} \tag{65a}$$

$$\lim_{z\to\infty} C(t,s) = 0 \tag{65b}$$

For the transfer function corresponding to B.C.(I), equation (60) is differentiated with respect to z:

$$\frac{\partial C(z,s)}{\partial z} = \frac{A_1\bar{u}}{2\bar{D}} e^{\frac{\bar{u}z}{2\bar{D}}(1+\beta')} + \frac{A_2\bar{u}}{2\bar{D}} e^{\frac{\bar{u}z}{2\bar{D}}(1-\beta')} \tag{66}$$

where $\beta' = \left[1 + (\frac{2\theta}{M})(s + k)\right]^{\frac{1}{2}}$

According to equation (62b), equation (66) is set to zero to solve for one constant in terms of another as:

$$A_1 = - \frac{A_2(1-\beta') e^{M(1-\beta')}}{(1+\beta') e^{M(1+\beta')}} \tag{67}$$

where $M = \frac{\bar{u}L}{2\bar{D}}$

Using equation (62a), the Laplace transform of the input is obtained as:

$$C_1(0,s) = \frac{A_1}{2}(1-\beta') + \frac{A_2}{2}(1+\beta') \tag{68}$$

Substitution of equation (67) into equation (68) gives:

$$C_1(0,s) = \frac{A_2}{2}\left[\frac{(1+\beta')^2 e^{M(1+\beta')} - (1-\beta')^2 e^{M(1-\beta')}}{(1+\beta') e^{M(1+\beta')}}\right] \tag{69}$$

The Laplace transform of output is:

$$C(L,s) = A_1 e^{M(1+\beta')} + A_2 e^{M(1-\beta')} \tag{70}$$

Substitution of equation (67) into equation (70) gives:

$$C(L,s) = A_2 \left\{ \frac{\pm 2\beta' e^{M(1-\beta')}}{1+\beta'} \right\} \tag{71}$$

According to equation (61), the transfer function is the ratio of equations (71) to (67), which upon simplification gives equation (19) of text.

For the transfer function corresponding to B.C.(II), equation (63a) is substituted into equation (60) to obtain:

$$C_1(0,s) = A_1 + A_2 \tag{72}$$

Since the second condition of B.C.(II) is the same as that of B.C.(I), equation (67) still holds for B.C.(II), and substitution of equation (67) into (72) gives:

$$C_1(0,s) = A_2 \left[\frac{(1+\beta') e^{M(1+\beta')} - (1-\beta') e^{M(1-\beta')}}{(1+\beta') e^{M(1+\beta')}} \right] \tag{73}$$

From equation (61), the transfer function corresponding to B.C.(II) is the ratio of equations (71) to (73), which upon simplification gives equation (20) of text.

For the transfer function corresponding to B.C.(III), equation (64b) is substituted to equation (60) to obtain:

$$\lim_{z \to \infty} C(z,s) = 0 = A_1(\infty) + A_2(0) \tag{74a}$$

So A_1 must be zero in order to satisfy equation (64b).

$$A_1 = 0 \tag{74b}$$

Since the first set of B.C.(III) is the same as that of B.C.(II), equation (72) still holds for B.C.(III). Substitution of equation (74) into (72) becomes:

$$C_1(0,s) = A_2 \tag{75}$$

The Laplace transform of the output is:

$$C(L,s) = A_2 e^{M(1-\beta')} \tag{76}$$

The transfer function corresponding to B.C.(III) is the ratio of equations (76) to (75), which upon simplification, gives equation (21) of the text.

Since B.C.(IV) is the combination of the first set of B.C. (I) and the second set of B.C.(II), equations (68) and (74) still hold for this case. Substitution of equation (74) into (68) gives:

$$C_1(0,s) = \frac{A_2(1+\beta')}{2} \tag{77}$$

The transfer function corresponding to B.C.(IV) is the ratio of equations (76) to (77), which upon simplification, gives equation (22) of text.

APPENDIX III

Systematic Derivations of The Response Functions of The Exit Concentrations Against Impulse Function and Step Inputs.

For the response functions of the exit concentration against impulse, it is known by L'Hospital rule that

$$C(L,s) = C_1 W(s) \tag{78}$$

For the solution corresponding to B.C.(I), equation (19) is substituted into equation (78) to obtain:

$$C(L,s) = C_1 \frac{4\beta'}{(1+\beta')^2 e^{-M(1-\beta')} - (1-\beta')^2 e^{-M(1+\beta')}} \tag{79}$$

The inverse transformation of equation (79) is carried out by means of the Heaviside partial fraction theorem. Since equation (79) is in the form of:

$$C(L,s) = \frac{q_i(s)}{p_i(s)} \tag{80}$$

where $q_1(s) = 4C_1\beta'$

$$p_1(s) = (1+\beta')^2 e^{-M(1-\beta')} - (1-\beta')^2 e^{-M(1+\beta')}$$

the inverse transform of equation (80) gives:

$$C(L,t) = \sum_{k=1}^{\infty} \frac{q_i(s_k)}{P_i'(s_k)} e^{s_k t} \tag{81}$$

where s_k is the root of $p_1(s) = 0$

Solving for the roots of $p_1(s) = 0$, one obtains:

$$s_k = - \frac{\delta n^2 + M^2}{2M\theta} - k \tag{82}$$

where δn = root of $\cot \delta = \frac{1}{2}\left(\frac{\delta}{M} - \frac{M}{\delta}\right)$

Differentiation of $p_1(s)$ with respect to s gives:

$$p'(s) = - \frac{\theta}{M\alpha}\left[M \cos M\alpha + 2 \cos M\alpha - 2M\alpha \sin M\alpha \right. \tag{83}$$

$$\left. - 2\alpha \sin M\alpha - M\alpha^2 \cos M\alpha \right]$$

where $\alpha = j\beta'$

Substitution of equation (82) into the expressions of $p_1'(s)$ and $q_1(s)$ gives respectively:

$$q(s_k) = C_1 \frac{2 \delta_{ne}{}^{M}}{M} \tag{84}$$

$$p'(s_k) = - \frac{\theta}{\delta n}\left[M \cos \delta n + 2 \cos \delta n - 2 \delta n \sin \delta n \right. \tag{85}$$

$$\left. - \frac{2 \delta n}{M} \sin \delta n - \frac{\delta n^2}{M} \cos \delta n \right]$$

Substitution of equations (84) and (85) into equation (81) gives:

$$\frac{C(L,t)}{C_1} = \sum_{n=1}^{\infty} \frac{1}{M\cos\delta n + 2\cos\delta n - 2\delta n \sin\delta n - \frac{2\delta n}{M}\sin\delta n - \frac{\delta n^2}{M}\cos\delta n} \times \left(\frac{2\delta n e^M}{M}\right)\left(-\frac{\delta n}{\theta}\right) \exp\left[-\left(\frac{\delta n^2 + M^2}{2M\theta}\right)t - kt\right] \tag{86}$$

Multiplication of both numerator and denominator on the right-hand side of equation (86) by $(M \sin\delta n + \delta n \cos\delta n)$ gives:

$$\frac{C(L,t)}{C_1} = \sum_{n=1}^{\infty}\left(-\frac{2\delta n^2}{M}\right)\left(\frac{M\sin\delta n + \delta n\cos\delta n}{K}\right) \times \exp\left[-\left(\frac{\delta n^2 + M^2}{2M\theta}\right)t - kt\right] \tag{87}$$

where $K = (M\cos\delta n + 2\cos\delta n - 2\delta n\sin\delta n - 2\frac{\delta n}{M}\sin\delta n - \frac{\delta n^2}{M}\cos\delta n)$

$$\times (M\sin\delta n + \delta n\cos\delta n)$$

Simplification of K gives:

$$K = -\frac{\delta n}{M}(M^2 + \delta n^2 + 2M) \tag{88}$$

Substitution of equation (88) into (87) and simplication, gives:

$$\frac{C_2(L,t)\theta}{C_1} = \sum_{n=1}^{\infty} \frac{2\delta n(M\sin\delta n + \delta n\cos\delta n)}{(M^2 + \delta n^2 + 2M)} \times \exp\left[M - \left(\frac{\delta n^2 + M^2}{2M} + R\right)\tau\right] \tag{89}$$

where $\tau = \frac{t}{\theta}$

The left-hand side of equation (89) is transformed in terms of the amount of tracer injected, Q, and the volume of reactor, V, by the following relationship:

$$Q = \bar{u} \int_o^\lambda C(\lambda)\, d\lambda = C_1 \pi d_r^2 u \tag{90}$$

$$V = \pi d_r^2 L \tag{91}$$

where d_r = diameter of reactor.

Hence, substitution of equations (90) and (91) into the left-hand side of equation (89) gives the following relationship:

$$\frac{C_2(L,t)}{C_1} = \frac{C_2(L,t)}{C_1} \times \frac{L}{u} = \frac{C_2 \pi d_r^2 L}{C_1 \pi d_r^2 u} = \frac{C_2 V}{Q} \tag{92}$$

Using equation (92) in place of equation (89) gives equation (23) of text.

For the solution corresponding to B.C.(II), equation (20) is substituted into equation (78) to obtain:

$$C(L,s) = C_1 \frac{2\beta'}{(1+\beta')\, e^{-M(1+\beta')} - (1-\beta')\, e^{-M(1-\beta')}} \tag{93}$$

Equation (93) is in the same form as equation (80), except $p_1(s)$ and $q_1(s)$ are replaced by the following two expressions:

$$q_2(s) = 2C_1\beta'$$

$$p_2(s) = (1+\beta')\, e^{-M(1+\beta')} - (1-\beta')\, e^{-M(1-\beta')}$$

Solving for the root of $p_2(s) = 0$, one obtains:

$$s_k = -\frac{\delta_n^2 + M^2}{2M\theta} - k \tag{94}$$

where δ'_n = root of $\tan\delta' = -\frac{\delta'}{M}$

Differentiation of p_2 with respect to s gives:

$$p_2'(s) = \left(-\frac{\theta}{M\alpha}\right)(M\cos M\alpha + \cos M\alpha - M\alpha \sin M\alpha) \tag{95}$$

Substitution of equation (94) into the expressions of $p_2'(s)$ and $q_2(s)$ gives respectively:

$$q_2(s_k) = \frac{\delta'_n}{M} e^M \tag{96}$$

$$p_2'(s_k) = -\left(\frac{\theta}{\delta'_n}\right)(M\cos\delta'_n + \cos\delta'_n - \delta'_n \sin\delta'_n) \tag{97}$$

Substitution of equations (96) and (97) into equation (81) gives:

$$\frac{C_2(L,t)}{C_1} = \sum_{n=1}^{\infty}\left(-\frac{\delta_n^2}{M}\right)\frac{1}{K_2}\exp\left[M - \left(\frac{\delta_n^2 + M^2}{2M\theta}\right)t - kt\right] \tag{98}$$

where $K_2 = M\cos\delta'_n + \cos\delta'_n - \delta'_n \sin\delta'_n$

Simplification of K_2 gives:

$$K_2 = \frac{M^2 + M + \delta_n^2}{M} \cos \delta'_n \tag{99}$$

Substitution of equation (99) into (98), upon simplification, gives:

$$\frac{C_2(L,t)\theta}{C_1} = \sum_{n=1}^{\infty} \frac{-\delta_n^2}{(M^2 + M + \delta'^2_n) \cos \delta'_n} \times \exp\left[M - \left(\frac{\delta_n^2 + M^2}{2M} + R\right)\tau\right] \tag{100}$$

Substitution of equations (90) and (91) into the left-hand side of equation (100) gives equation (24) of text.

For the solution corresponding to B.C.(III), equation (21) is substituted into equation (78) to obtain:

$$C(L,s) = C_1 \exp\left[M(1-\beta')\right] \tag{101}$$

If one lets $g_1 = L/\sqrt{\bar{D}}$ and $g_2 = \frac{\bar{u}^2}{4\bar{D}} + k$, equation (101) is simplified to:

$$C(L,s) = C_1 e^M e^{-g_1\sqrt{g_2 + s}} \tag{102}$$

Equation (102) is transformed to the following expression by the rule of "shift of original function" as:

$$\frac{C(L,t)}{C_1} = e^{M - g_2 t} \mathcal{L}^{-1}\left\{e^{-g_1\sqrt{s}}\right\} \tag{103}$$

The inverse transformations of equation (103) is carried out by means of tables of integral transforms (5):

$$\mathcal{L}^{-1}\left\{e^{-g_1\sqrt{s}}\right\} = \frac{g_1}{2\sqrt{\pi t^3}} \exp\left(-\frac{g_1^2}{4t}\right) \qquad (104)$$

Substitution of g_1 and g_2 into equation (104) gives:

$$\frac{C_2\theta}{C_1} = \sqrt{\frac{M}{2\pi}}\ (\tau)^{-\frac{3}{2}} \exp\left[M - \tfrac{1}{2}\left(M\tau + \frac{M}{\tau}\right) - R\tau\right] \qquad (105)$$

Use of equations (90) and (91) in place of the left-hand side of equation (105) gives equation (25) of text.

For the solution corresponding to B.C.(IV), equation (22) is substituted into equation (78) to obtain:

$$C(L,s) = C_1\ \frac{2\exp\left[M(1-\beta')\right]}{(1+\beta')} \qquad (106)$$

If one lets $g_3 = \frac{L}{\sqrt{D}}$, $g_4 = \frac{\bar{u}^2}{4D} + k$, and $g_5 = \frac{M}{g_3}$, equation (106) becomes:

$$\frac{C(L,s)}{C_1} = 2g_5\, e^{M}\left\{\frac{e^{-g_3\sqrt{g_4+s}}}{g_5+\sqrt{g_4+s}}\right\} \qquad (107)$$

The inverse transformations of equation (107) by means of "shifts of original function" and the tables of integral transforms (5) gives:

$$\frac{C(L,t)}{C_1} = 2g_5\, e^{M-g_4 t}\, \frac{1}{\sqrt{\pi t}} \exp\left(-\frac{g_3^2}{4t}\right) - g_5 \exp(g_3 g_5 + g_5^2 t)$$

$$\times \operatorname{Erfc} \frac{g_3}{2\sqrt{t}} + g_5\sqrt{t} \qquad (108)$$

Substitution of g_1, g_2 and g_3 into equation (108), upon simplification, gives equation (26) of text.

The response functions of the exit concentration against step input are known to be:

$$C(L,s) = \frac{C_1 W(s)}{s} \qquad (109)$$

Since the transfer functions, W(s), corresponding to each of four boundary conditions and their inverse transforms are known by the derivations of response functions against impulse, the following relationships can be expressed from equation (109) by the method of convolution:

$$\frac{C(L,t)}{C_1} = \int_0^{\tau} \sum_{n=1}^{\infty} \frac{2\delta_n(M\sin\delta_n + \delta_n\cos\delta_n)}{(M^2 + 2M + \delta_n^2)}$$

$$\times \exp\left[M - \left(\frac{\delta_n^2 + M^2}{2M} + R\right)\lambda\right] d\lambda \qquad (110)$$

$$\frac{C(L,t)}{C_1} = \int_0^{\tau} \sum_{n=1}^{\infty} \frac{-\delta'^2_n}{(M^2 + M + \delta'^2_n)\cos\delta'_n}$$

$$\times \exp\left[M - \left(\frac{\delta'^2_n + M^2}{2M} + R\right)\lambda\right] d\lambda \qquad (111)$$

$$\frac{C(L,t)}{C_1} = \int_0^{\tau} \sqrt{\frac{M}{2\pi}} (\lambda)^{-\frac{3}{2}} \times \exp\left[M - \tfrac{1}{2}\left(M\lambda + \frac{M}{\lambda} + 2\lambda R\right)\right] d\lambda \tag{112}$$

$$\frac{C(L,t)}{C_1} = \int_0^{\tau} 2\left\{\exp\left[M-(M+2R)\frac{\lambda}{2}\right] \operatorname{Erfc}\sqrt{\frac{M}{2\lambda}} - \exp(2M - R\lambda) \operatorname{Erfc}\left(\sqrt{\frac{M}{2\lambda}} + \sqrt{\frac{M\lambda}{2}}\right)\right\} d\lambda \tag{113}$$

The integration of equations (110) and (111) are straight forward, and their results can be obtained easily as shown by equations (27) and (28) of the text respectively. The integrations of equations (112) and (113) are difficult to carry out by the method of convolution because they contain the error functions. Heaviside's partial fraction theorem was found to be more convenient for the inverse transform of these two equations.

For the solution corresponding to B.C.(III), equation (21) is substituted into equation (109) to obtain:

$$C(L,s) = \frac{C_1 e^{M(1-\beta')}}{s} \tag{114}$$

If one lets $g_6 = \frac{L}{\sqrt{\bar{D}}}$ and $g_7 = \frac{\bar{u}}{4\bar{D}} + k$, equation (114) is simplified to:

$$\frac{C(L,s)}{C_1} = \frac{e^{-g_6\sqrt{g_7 + s}}}{s} \cdot e^{M} \tag{115}$$

Equation (115) is transformed to the following expression by the rule of "shift of original function" as:

$$\frac{C(L,t)}{C_1} = e^{M-g_7 t}\, \mathcal{L}^{-1}\left\{ \frac{e^{-g_6\sqrt{s}}}{s} \right\} \tag{116}$$

The inverse transformation of equation (116) is carried out by means of the table of integral transforms (5):

$$\frac{C(L,t)}{C_1} = \frac{1}{2}\left\{ e^{M-(-g_6\sqrt{g_7})}\ \mathrm{Erfc}\left(\frac{g_6}{2\sqrt{t}} - \sqrt{g_7 t} \right) \right.$$
$$\left. + e^{M+(-g_6\sqrt{g_7})}\ \mathrm{Erfc}\left(\frac{g_6}{2\sqrt{t}} - \sqrt{g_7 t} \right) \right\} \tag{117}$$

Substitution of g_6 and g_7 into equation (117) gives equation (29) of text.

The analytical solution corresponding to B.C.(IV) is found to be difficult because it contains the error function and can not be integrated by the method of convolution as shown in equation (113). The solution for a step input corresponding to B.C.(IV) is left in integral form as shown in equation (30) of the text. An approximate method, however, such as integration in a series form could be carried out by expanding the error functions.

APPENDIX IV

Derivations of Frequency Response Functions In Terms of Amplitude Ratios and Phase Shift.

Since the performance functions are defined as the ratio of Fourier transforms of output to input, they are systematically obtained from the transfer functions by replacing s by $j\omega$ as:

$$P.F. = \frac{4\beta''}{(1+\beta'')^2 \exp\left[-M(1-\beta'')\right] - (1-\beta'')^2 \exp\left[-M(1+\beta'')\right]} \qquad (118)$$

$$P.F. = \frac{2\beta''}{(1+\beta'') \exp\left[-M(1-\beta'')\right] - (1-\beta'') \exp\left[-M(1+\beta'')\right]} \qquad (119)$$

$$P.F. = \exp\left[M(1-\beta'')\right] \qquad (120)$$

$$P.F. = \frac{2 \exp\left[M(1-\beta'')\right]}{(1+\beta'')} \qquad (121)$$

where $\beta'' = \sqrt{1 + \left(\frac{2\theta}{M}\right)\left(j\omega + k\right)}$

The complex number, β'', of equations (118) through (121) is arranged in polar form as:

$$\beta'' = \left[1 + \frac{2R}{M} + \frac{2(\omega\theta)}{M}\right] = \left[\left(1 + \frac{2R}{M}\right)^2 + \frac{4(\omega\theta)^2}{M^2}\right]^{\frac{1}{4}} e^{ja} \qquad (122)$$

where $a = \frac{1}{2} \operatorname{Tan}^{-1}\left(\frac{2\omega\theta}{2R + M}\right)$

For the solution corresponding to B.C.(I), equation (122) is substituted to obtain:

$$\frac{C_2(j\omega)}{C_1(j\omega)} = \frac{4\beta'' e^{M}(\cos a + j\sin a)}{4\left[1+\frac{(\omega\theta)}{M}j\right]\sinh(x+jy) + 4b(\cos a+j\sin a)\cosh(x+jy)} \qquad (123)$$

Separation of the denominator in equation (123) into a real part and an imaginary part gives:

$$\frac{C_2(j\omega)}{C_1(j\omega)} = \frac{4be^{M}(\cos a + j \sin a)}{4(\alpha_1 + j\alpha_2)} \qquad (124)$$

where α_1 and α_2 are given in the text.

Equation (124) is, then transformed into the polar form as:

$$\frac{C_2(j\omega)}{C_1(j\omega)} = \frac{be^{M}}{(\alpha_1^2 + \alpha_2^2)^{\frac{1}{2}}} \exp\left\{j\left[a-\mathrm{Tan}^{-1}\left(\frac{\alpha_2}{\alpha_1}\right)\right]\right\} \qquad (125)$$

Transformation of equation (125) into the amplitude ratio and phase shift results in equation (31) of the text.

For the solution using B.C.(II), equation (122) is substituted into equation (119) which, upon simplification, gives:

$$\frac{C_2(j\omega)}{C_1(j\omega)} = \frac{2be^{M}(\cos a + j\sin a)}{\left\{1+b(e^{ja})\right\}\exp(x+jy) - \left\{1-b(e^{ja})\right\}\exp[-(x+jy)]} \qquad (126)$$

Further separation of the denominator in equation (126) into real and imaginary part gives:

$$\frac{C_2(j\omega)}{C_1(j\omega)} = \frac{2be^M (\cos a + j \sin a)}{2(\alpha_3 + \alpha_4)} \qquad (127)$$

where α_3 and α_4 are given in the text.

Rearrangement of equation (127) into the polar form gives:

$$\frac{C_2(j\omega)}{C_1(j\omega)} = \frac{be^M}{(\alpha_3^2 + \alpha_4^2)^{\frac{1}{2}}} \exp\left\{ j\left[a - \tan^{-1}\left(\frac{\alpha_4}{\alpha_3}\right)\right]\right\} \qquad (128)$$

Equation (128) is then transformed into amplitude ratio and phase shift, which give equation (32) of the text.

Substitution of equation (122) into equation (120) gives the solution for B.C.(III) as:

$$\frac{C_2(j\omega)}{C_1(j\omega)} = \exp M\left[1-b(\cos a + j \sin a)\right] = e^{M-x} e^{-jy} \qquad (129)$$

Equation (33) of the text is obtained by transforming equation (129) in terms of amplitude ratio and the phase shift.

For the solution using B.C.(IV), equation (122) is substituted into equation (121), which upon simplification, gives:

$$\frac{C_2(j\omega)}{C_1(j\omega)} = 2\, e^{M-x}\left[\frac{\cos y - j \sin y}{\left(1 + \frac{x}{M}\right) + j(\sin a)}\right] \qquad (130)$$

Rearrangement of equation (130) in polar form gives:

$$\frac{C_2(j\omega)}{C_1(j\omega)} = \frac{2\, e^{M-x}}{\left[\left(1 + \frac{\quad}{\quad}\right)^2 + \sin^2 a\right]^{\frac{1}{2}}} \exp\left[-y - \mathrm{Tan}^{-1}\left(\frac{\sin a}{1 + \frac{x}{M}}\right)\right] \qquad (131)$$

Equation (131) is then transformed into amplitude ratio and phase shift which give equation (34) of the text.

A DIFFUSION MODEL OF AN ISOTHERMAL TUBULAR FLOW REACTOR

by

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The general extent of axial dispersion and its effect on the performance of isothermal tubular flow reactors are investigated by use of the diffusion-type model and four sets of following boundary conditions:

$$\text{B.C.(I)} \quad \begin{cases} \bar{u}C_1 = \bar{u}C_{z\to 0^+} - \bar{D}\left(\dfrac{\partial C}{\partial z}\right)_{z\to 0^+} \\ \left(\dfrac{\partial C}{\partial z}\right)_{z=L} = 0 \end{cases}$$

$$\text{B.C.(II)} \quad \begin{cases} \bar{u}C_1 = \bar{u}C_{z\to 0^+} \\ \left(\dfrac{\partial C}{\partial z}\right)_{z=L} = 0 \end{cases}$$

$$\text{B.C.(III)} \quad \begin{cases} \bar{u}C_1 = \bar{u}C_{z\to 0^+} \\ \lim\limits_{z\to\infty} C(z,t) = 0 \end{cases}$$

$$\text{B.C.(IV)} \quad \begin{cases} \bar{u}C_1 = \bar{u}C_{z\to 0^+} - \bar{D}\left(\dfrac{\partial C}{\partial z}\right)_{z\to 0^+} \\ \lim\limits_{z\to\infty} C(z,t) = 0 \end{cases}$$

The differences and the relationship among the steady-state concentration profiles and conversion and unsteady-state transient response curves obtained by use of the different sets of boundary conditions are critically re-examined and numerically evaluated. Only the values obtained by use of B.C.(I) are found to be consistent with the limits of maximum and minimum conversions, i.e. conversion in a plug-flow reactor and in a complete-

ly stirred tank reactor calculated from a one-dimensional transport equation. The solutions obtained by use of the other three sets of boundary conditions are found to be inconsistent with such limits. Hence, unless their limits are recognized, the use of B.C.(II), (III) and (IV) should be avoided. Due to the simple analytical solutions of equations in exponential form, the use of B.C.(III) and (IV), however, offers a convenient means for computing final conversions if their limits are recognized.

An illustration is presented of the method of determining mean dispersion coefficients from dynamic experiments. It is recommended that the calculated values should be compared with the experimental data to determine the feasibility of predicted models and boundary conditions.

The response curves based on different types of transient inputs are presented for all four different sets of boundary conditions both with and without chemical reaction. These curves should be useful for practicing process control engineers and for those who wish to extract the transport parameter from dynamic response experiments.